

Review

A systematic review of valuable resource recovery from spent lithium-ion battery cathodes: Technologies, challenges, and future trends

Jianqiu Qin^{1,4}, Shaoying Li^{1,2,3,4,*} , Beining Liu¹, Xiaodong Yu¹, Liucheng Zhao^{1,3,4,*}

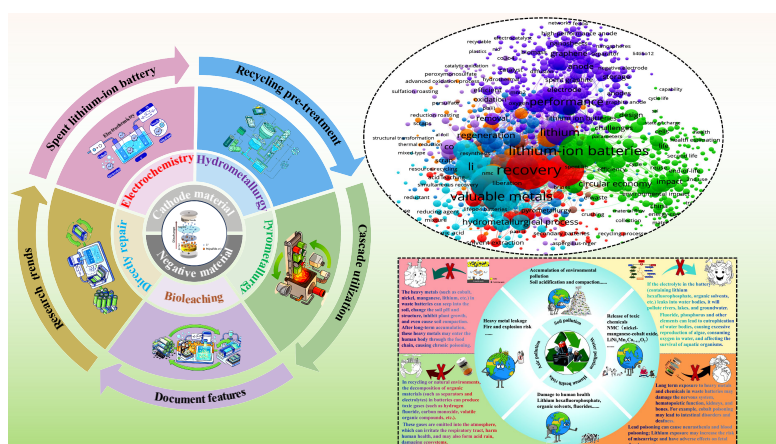
¹ College of Mining Engineering, North China University of Science and Technology, Tangshan 063210, China

² State Key Laboratory of Mineral Processing, Beijing 100160, China

³ Collaborative Innovation Center for Green Development and Ecological Restoration of Mineral Resource, Tangshan 063210, China

⁴ Hebei Mining Development and Safety Technology Laboratory, Tangshan 063210, China

Abstract: This review systematically summarizes the current research status and development trends in the recovery of valuable resources from spent lithium-ion battery (SLIB) cathodes, aiming to elucidate key technological pathways, research hotspots, and future challenges in this field. By conducting a bibliometric analysis of the field of valuable resource recovery from SLIBs, combined with an in-depth analysis of pretreatment, cascade utilization, and various regeneration technologies—including hydrometallurgy, pyrometallurgy, bioleaching, direct regeneration, electrochemical methods, and ionic liquid-based recovery—this review reveals that although significant progress has



been made in current recovery processes, they still generally face challenges such as high energy consumption, low metal recovery efficiency, severe secondary pollution, and difficulties in scaling up. Future recovery technologies for valuable resources from SLIBs should be oriented towards green and low-carbon development, intelligent systems, and systematic approaches. Through multi-technology integration, process optimization, and the establishment of closed-loop systems, it is possible to achieve efficient recovery of critical metals and sustainable management of the entire battery life cycle. This review is expected to provide systematic solutions for resource recycling and environmental protection.

Keywords: spent lithium-ion batteries, recycling, hydrometallurgy, pyrometallurgy, bioleaching, direct regeneration, electrochemical recovery

Citation: J. Qin, S. Li, B. Liu, X. Yu, L. Zhao. A systematic review of valuable resource recovery from spent lithium-ion battery cathodes: Technologies, challenges, and future trends. *Materials Reports: Solidwaste and Ecomaterials*, 2026, 2: 9520029. <https://doi.org/10.26599/MRSE.2026.9520029>.

1. Introduction

Driven by the global transition towards a cleaner and low-car-

bon energy mix, the demand for lithium-ion batteries (LIBs)^[1-4], serving as pivotal energy storage solutions for elec-

*Correspondence to S. Li, lishaoying@ncst.edu.cn; L. Zhao, zhaoliucheng@ncst.edu.cn

Received 14 May 2026; Received in revised form 12 June 2026; Accepted 28 June 2026

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tric vehicles, portable electronic devices, and large-scale energy storage systems, has been experiencing a sustained and dramatic surge^[5-7]. However, the practical service life of LIBs is limited^[8-10]. Amidst the rapid expansion of the electric vehicle industry and the accelerated iteration of electronic products, the world is on the brink of a massive wave of battery retirements^[11-13]. If not properly managed, the toxic transition metals (e.g., cobalt, nickel, manganese), organic electrolytes, and fluorinated phosphorus compounds contained in spent batteries pose significant threats to ecological systems and human health. In addition, although lithium itself is not classified as a heavy metal, its improper disposal can still lead to soil and water contamination, necessitating careful management^[14-16], as illustrated in Fig. 1. Conversely, the high abundance of critical metal elements such as lithium^[17-19], cobalt, nickel, and manganese in these retired batteries represents substantial resource value^[20-22], positioning them as indispensable secondary strategic resources. There-

fore, developing efficient, green, and sustainable recycling technologies for recovering valuable resources from spent LIBs is not only a critical pathway for achieving resource circularity and alleviating supply pressure on primary mineral resources^[23-25], but also constitutes a strategic initiative for promoting the green and low-carbon transformation of the battery industry chain^[26-28], mitigating environmental and health risks, and establishing a closed-loop circular economy^[29-31].

In recent years, numerous review studies have been dedicated to the resource-oriented recycling of spent lithium-ion batteries (SLIBs), systematically summarizing the advancements and challenges associated with conventional recycling technologies such as hydrometallurgy, pyrometallurgy, and bioleaching^[32-35]. However, the current review studies in this field still exhibit notable limitations, as illustrated in Fig. 2, which constrain their utility in guiding both academic research and engineering practices. First, while most studies have focused on a single or a limited number of mainstream

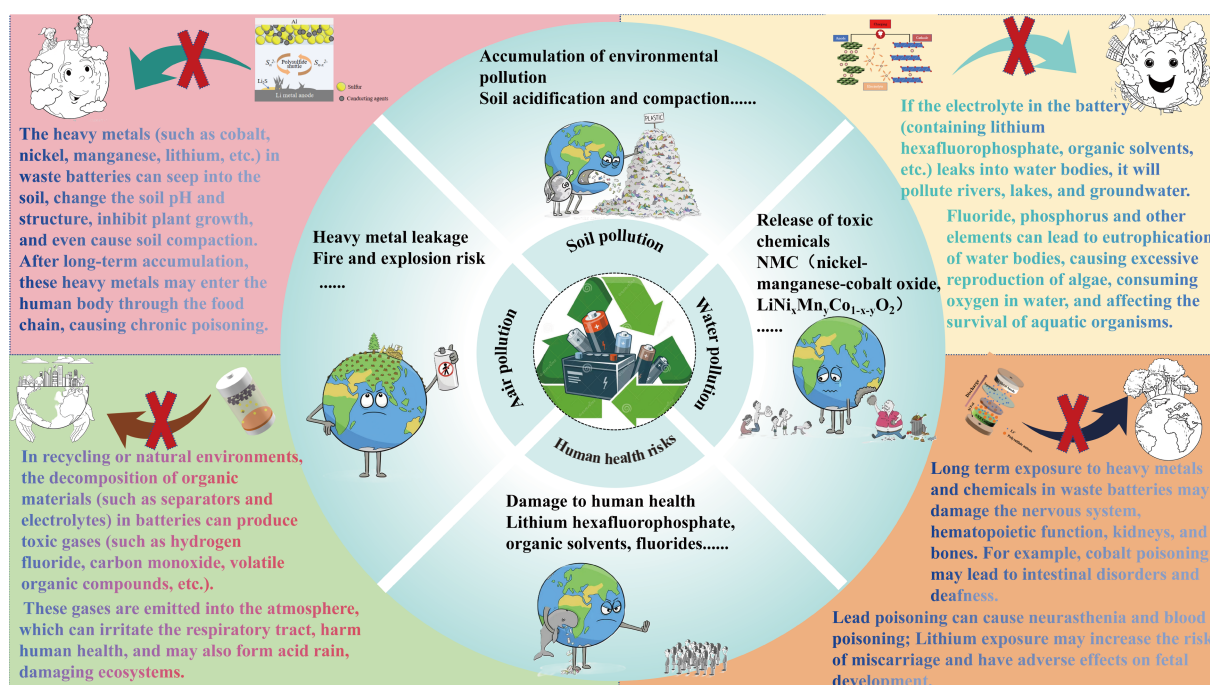


Fig. 1. Environmental and health hazards associated with improper handling of spent lithium-ion batteries.

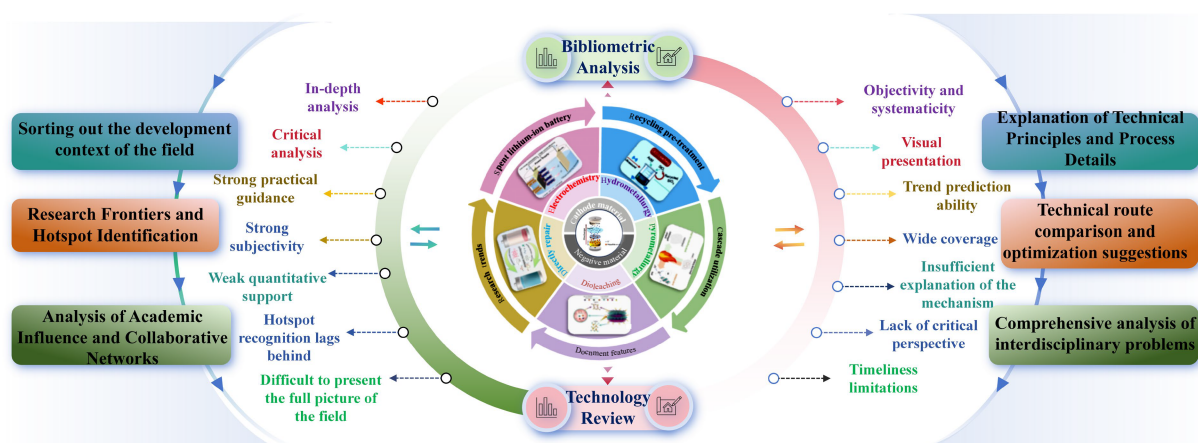


Fig. 2. Comparison of advantages and disadvantages between bibliometric analysis and traditional review methods.

processes, there is a lack of comprehensive reviews systematically addressing emerging green technologies such as direct regeneration and repair, electrochemical recycling, and the use of ionic liquids and deep eutectic solvents (DES). This gap highlights the need for more comparative and synergistic analysis across multiple technological pathways. Second, existing reviews predominantly rely on qualitative descriptions and have yet to fully integrate bibliometric and scientometric mapping tools. This oversight hinders the quantitative and visual analysis of the field's development trends, the evolution of research hotspots, and collaborative networks, ultimately resulting in an incomplete and less objective grasp of the discipline's developmental trajectory. To address these research gaps, this paper adopts an integrated research framework that combines bibliometric analysis with in-depth reviews of multiple technological pathways, aiming to systematically consolidate the research progress in the field of valuable resource recovery from SLIBs. By integrating bibliometric analysis with in-depth multi-technology pathway assessments, a tripartite research framework of "data-mechanism-system" has been constructed. This approach enables a holistic analysis spanning from the macro-level landscape of the field to the micro-level technical bottlenecks, and further proposes synergistic innovation pathways for the green, intelligent, and systematic development of SLIB recycling^[36-38].

This paper aims to construct a systematic knowledge

framework that integrates macro-level trends, technological mechanisms, and cutting-edge advancements. It seeks to provide a reference for academic research, engineering applications, and industrial decision-making in the field of SLIB resource recovery, underpinned by both data-driven insights and theoretical depth. The ultimate goal is to propel recycling technologies towards higher efficiency, greener processes, and greater intelligence, thereby contributing to the synergistic achievement of a global battery circular economy and sustainable development goals.

2. Bibliometrics and research hotspot analysis

2.1. Analysis of publication characteristics and collaboration networks

Drawing upon the relevant literature from the Web of Science (WOS) core collection, this review employs CiteSpace and VOSviewer to conduct a scientometric analysis of the research field dedicated to SLIB recycling technologies, as illustrated in Fig. 3. The annual publication volume in this field has grown steadily, with a notable increase after 2019. This trend reflects the growing research interest driven by policy support and the rapid expansion of the electric vehicle market. However, bibliometric analysis alone cannot distinguish between incremental contributions and transformative advances. A more meaningful assessment requires examin-

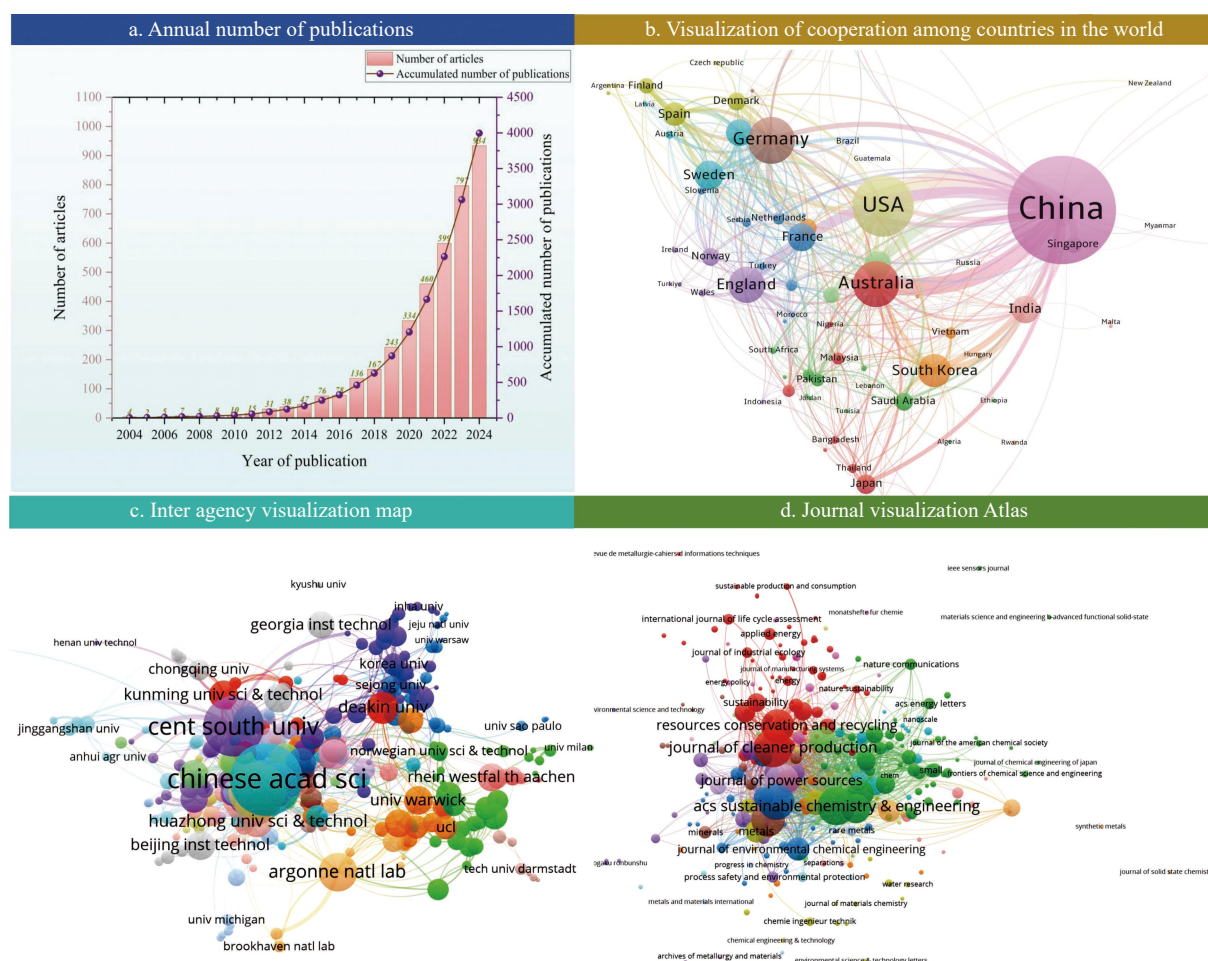


Fig. 3. Analysis of publication characteristics and collaboration network in the research field of spent lithium-ion battery recycling technologies.

ing citation impact, research diversity, and the extent to which laboratory findings translate into industrial applications. Analysis of national and institutional contributions shows that China, the United States, and South Korea are the most active countries in this research area, with Chinese institutions accounting for a substantial share of total publications. However, the inter-institutional collaboration network remains relatively sparse, suggesting that research capacity is concentrated within a limited number of institutions. Notably, high publication output does not necessarily correlate with high citation impact. Future assessments should consider metrics such as field-weighted citation impact and the diversity of research topics addressed. Co-authorship analysis reveals an interconnected network of researchers, with several authors contributing to multiple publications. Co-citation analysis further identifies key researchers whose work has been frequently referenced, such as Li Li, whose studies on direct regeneration and closed-loop recycling have shaped subsequent research directions. Rather than focusing on publication counts, the analysis highlights the emergence of research clusters centered around specific technical themes, including hydrometallurgical process optimization, direct material regeneration, and life cycle assessment. Journal analysis reveals that the *Journal of Cleaner Production* and *ACS Sustainable Chemistry & Engineering* are among the primary outlets for SLIB recycling research, reflecting the field's emphasis on sustainability and green chemistry. The *Journal of Power Sources*, with its long-standing focus on battery technology, ranks among the most cited journals, indicating its foundational role in the field. Beyond publication counts, the diversity of journal sources—spanning chemical engineering, environmental science, and materials science—underscores the interdisciplinary nature of this research area. Co-citation analysis of references further delineates key review articles and foundational studies, highlighting prominent research themes such as the greening of recycling technologies, systematic process optimization, and sustainability assessments. The research landscape for SLIB recycling has formed a multinational collaborative framework centered on China and the United States. However, the long-term impact of this research will be determined not by publication counts, but by the extent to which laboratory-scale innovations translate into industrially scalable, economically viable, and environmentally sustainable recycling processes. Future assessments should prioritize metrics that capture research influence, such as citation impact, technology readiness levels, and successful industry partnerships. Publication outputs have been concentrated in high-impact journals such as the *Journal of Cleaner Production* and *ACS Sustainable Chemistry & Engineering*, with citation metrics indicating broad academic influence within the field. This field is anticipated to continue its deepening trajectory, driven by the dual forces of policy support and market dynamics.

To advance the green and sustainable development of SLIB recycling technologies, future research and practical efforts should prioritize the following directions: Firstly, international collaboration networks should be expanded to facilitate the global flow and sharing of research outcomes and cut-

ting-edge technologies. Secondly, in-depth interdisciplinary and cross-institutional cooperation should be promoted, integrating expertise from materials science, environmental engineering, and policy management. Furthermore, efforts should focus on developing green, low-carbon, and cost-effective recycling processes to enhance the recovery efficiency of critical metals and the comprehensive performance of regenerated materials. Finally, it is essential to establish an integrated management system covering the entire battery life cycle, promote the implementation of standardized recycling models, and strengthen the systematic evaluation of their environmental and economic benefits.

2.2. Research dynamics and hotspot analysis

This review systematically examines the research dynamics within the field of SLIB recycling technologies, identifying several core research themes and their evolutionary trajectories, as illustrated in Fig. 4. Current research is predominantly centered on several key themes, including battery performance assessment, the recovery of critical metals, material regeneration technologies, and the comprehensive optimization of environmental and economic benefits. Specifically, the recovery and reuse of high-performance electrode materials (such as lithium- and nickel-based cathode materials) is a focus of research, and its technical path covers hydrometallurgy, direct regeneration of materials, etc. At the same time, the recovery and treatment of graphite anode, electrolyte, and other components are also gradually being paid attention to, which shows that the field is expanding from single metal recovery to full component recycling. From the perspective of the time dimension, research hotspots show obvious characteristics of phased evolution. Around 2015, with the rapid rise of the new energy vehicle industry, the research focused on the nano and composite modification of electrode materials. The highlight of keywords such as “nanoparticles” and “composite” reflects the urgent demand for high-performance electrode materials in this period. In recent years (2022–2024), the research frontier has been further deepened, showing three significant changes: first, the shift from laboratory scale to engineering application, with keywords such as “black mass” and “system” reflecting the concern for large-scale processing technology; second, the extension from single technology development to full life cycle management, where the wide application of “material flow analysis” and other methods marks the formation of a systematic research paradigm; third, the transition from traditional pyrometallurgy to green low-carbon processes, where the exploration of new recovery routes such as “carbothermal reduction” reflects the urgent need of the industry to reduce the carbon footprint. These trends show that the field of SLIB recycling is forming a complete interdisciplinary research system covering materials science, process engineering, system analysis, and other disciplines.

Based on the above research status and trend analysis, future research and practice in the field of SLIB recycling should focus on the following three directions. First, efforts should be made to develop green and low-carbon closed-loop recycling processes. At present, the recovery process still faces problems such as high energy consumption and sec-

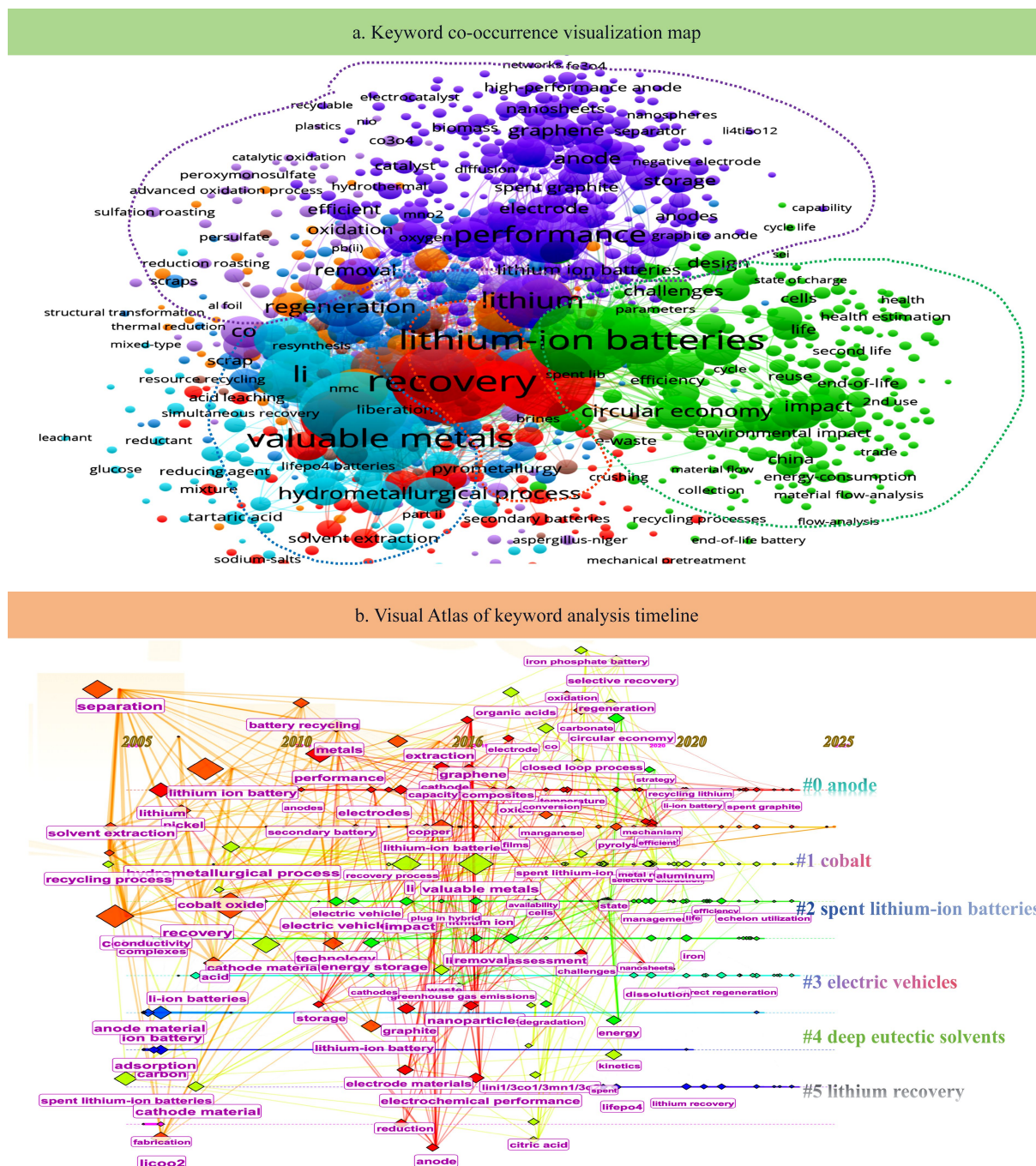


Fig. 4. Analysis of research dynamics and hotspots in the research field of spent lithium-ion battery recycling technologies.

ondary pollution risk. In the future, the development needs to focus on breaking through low-energy and low-emission technologies such as low-temperature metallurgy, bioleaching, and selective electrolysis, while strengthening the harmless treatment and resource utilization of non-metallic components such as electrolyte and graphite, and building a full-chain closed-loop system of “recovery-regeneration-reuse”. Second, the upgrading of recycling processes should focus on improving automation and precision in unit operations such as disassembly and sorting. While machine vision and learning-based algorithms have been explored in laboratory settings, their industrial deployment remains constrained by the high variability of spent battery feedstocks, the lack of labeled training data, and the difficulty of integrating such sys-

tems into existing processing lines. Substantial engineering validation and domain-specific adaptation are required before these tools can deliver reliable gains in operational efficiency. Third, the application of material flow analysis and life cycle assessment methods should be deepened, a carbon footprint tracking system covering the whole life cycle of batteries should be established, and data support and system optimization schemes should be provided for the green and low-carbon transformation of the recycling process. Finally, we should strengthen the interdisciplinary integration and collaborative innovation of the industrial chain. The in-depth integration of materials science, chemical engineering, environmental management, and other disciplines should be promoted to overcome the core technology bottlenecks such as direct

regeneration of electrode materials and short-range recovery of key metals. At the same time, a collaborative innovation platform of “industry-university-research-application” should be established to promote the international docking of recycling technology and industrial standards, and provide systematic solutions for building a sustainable battery recycling economy system.

3. Current research landscape and challenges

3.1. Pretreatment technologies for SLIB recycling

The efficient pretreatment of SLIBs is regarded as the key link for achieving resource recovery and environmental risk control, and plays a decisive role in the subsequent material separation and high-value utilization^[39,40]. The standardized basic procedure consists of discharging, disassembly, crushing, and sorting. Discharging eliminates residual electricity to avoid safety hazards. Disassembly separates the battery shell from its internal structure. Crushing increases the contact area for subsequent treatment. Sorting enables the separation of metals, plastics, and electrode materials^[41]. The core objectives are defined as achieving effective separation of components, eliminating the risk of hazardous substance leakage, and enriching target recyclable materials such as cathode active materials, thereby improving the overall economics of subsequent recycling processes^[42]. Currently, pretreatment technologies are mainly categorized into physical and chemical methods. Physical pretreatment techniques, widely adopted due to their environmental compatibility and operational simplicity, include mechanical processing, mechanochemical treatment, and thermal methods. It should be noted that while solvent dissolution is sometimes conceptually associated with physical routes, it inherently relies on chemical interactions (e.g., polarity matching and solvation) and is therefore more appropriately classified as a chemical method. Mechanical methods primarily involve crushing and magnetic separation. Mechanochemical methods include ball milling technology and chemically induced reactions. Thermal methods are represented by high-temperature calcination and organic decomposition. Component separation in physical pretreatment is achieved through mechanical force, mechanochemical reactions induced by mechanical energy, and temperature regulation^[43,44]. Chemical pretreatment technology, in contrast, relies on chemical reactions such as acid leaching and alkaline leaching to achieve efficient separation and purification. Compared to physical methods, chemical pretreatment offers higher selectivity and metal recovery rates but requires more complex reagent handling and generates wastewater. Physical pretreatment is simpler and cleaner from an operational standpoint but often yields lower product purity and less effective impurity removal. The choice between physical and chemical pretreatment should therefore be guided by downstream processing requirements and environmental constraints. In acid leaching, metal ions are dissolved by the oxidation and complexation actions of acidic solutions, and the leaching rate is improved by optimizing parameters such as acidity and temperature. Alkaline leaching technology is used to dissolve and remove impurities from aluminum foil, enabling the enrichment of cathode active materials and demonstrat-

ing significant advantages in the selective recovery of target components^[45,46].

Kar et al.^[47] evaluated four process combinations for SLIB recycling pretreatment, involving discharge mode (NaCl solution and liquid nitrogen at low temperature), binder removal (N-methyl-2-pyrrolidone solvent and solvent-free) and grinding mode (normal temperature and low temperature), as illustrated in Fig. 5. By comparing the performance of each scheme in terms of operability, safety, energy consumption, material enrichment, and particle size distribution, and combining life cycle assessment and economic analysis, they found that the low-temperature solvent-free process had the best comprehensive performance. Although NaCl solution discharge was environmentally friendly, it was inefficient and had potential safety hazards. Li et al.^[48] developed a pyrolysis-ultrasound-micro-nanobubble flotation combined pretreatment process for recycling SLIBs. Firstly, the binder and electrolyte were removed by pyrolysis in an inert atmosphere (350 °C, 15 min), and then the residual carbon layer was stripped by ultrasonic or grinding. Finally, the grade of cathode material was increased from about 65% to 95% by micro-nanobubble flotation, and the recovery rate was increased from 66% to 88%. The surface properties and morphology changes of the electrode before and after pyrolysis were characterized and analyzed. The mechanism of micro-nanobubbles enhancing the adhesion of graphite through the “bubble bridge” mechanism was revealed. It was proved that this method was superior to the traditional wet process in terms of reagent consumption, energy consumption, and emission. Fang et al.^[49] revealed the low-temperature inerting mechanism of liquid nitrogen and dry ice: below −60 °C, the battery electrolyte solidifies, and the internal resistance increases sharply, resulting in the loss of output capacity. Acupuncture experiments show that the technology can suppress the peak temperature of thermal runaway from 921 °C to below 150 °C. The research points out that the cooling durability of dry ice is better than that of liquid nitrogen due to its high latent heat and low cost. Therefore, the proposed “freezing-crushing” process provides a key basis for safe recovery. Wu et al.^[50] verified a “deepening disassembly” preprocessing paradigm with robot and ultrasonic cutting as the core. This technology can automatically separate the electrode components, realize the accurate classification of materials from the source, improve the purity of active materials from 33.8% to 46.6%, and reduce aluminum impurities by two orders of magnitude, providing high-quality raw materials for direct regeneration. Li et al.^[51] revealed the reaction mechanism and kinetics of pyrolysis pretreatment of spent lithium iron phosphate (LFP) batteries in argon. The optimal pyrolysis window was 500–600 °C, and the defluorination rate was 91.16% without destroying the LFP crystal phase. Through the kinetic model, the reaction follows three-dimensional diffusion control, and the apparent activation energy is 123 kJ mol^{−1}, which provides a theoretical basis for process optimization. Two recent reviews have addressed human-robot collaborative disassembly (HRCD) in battery recycling, yet with distinct emphases. Li et al.^[52] focus on artificial intelligence (AI) and human-robot collaboration (HRC) in retired battery disas-

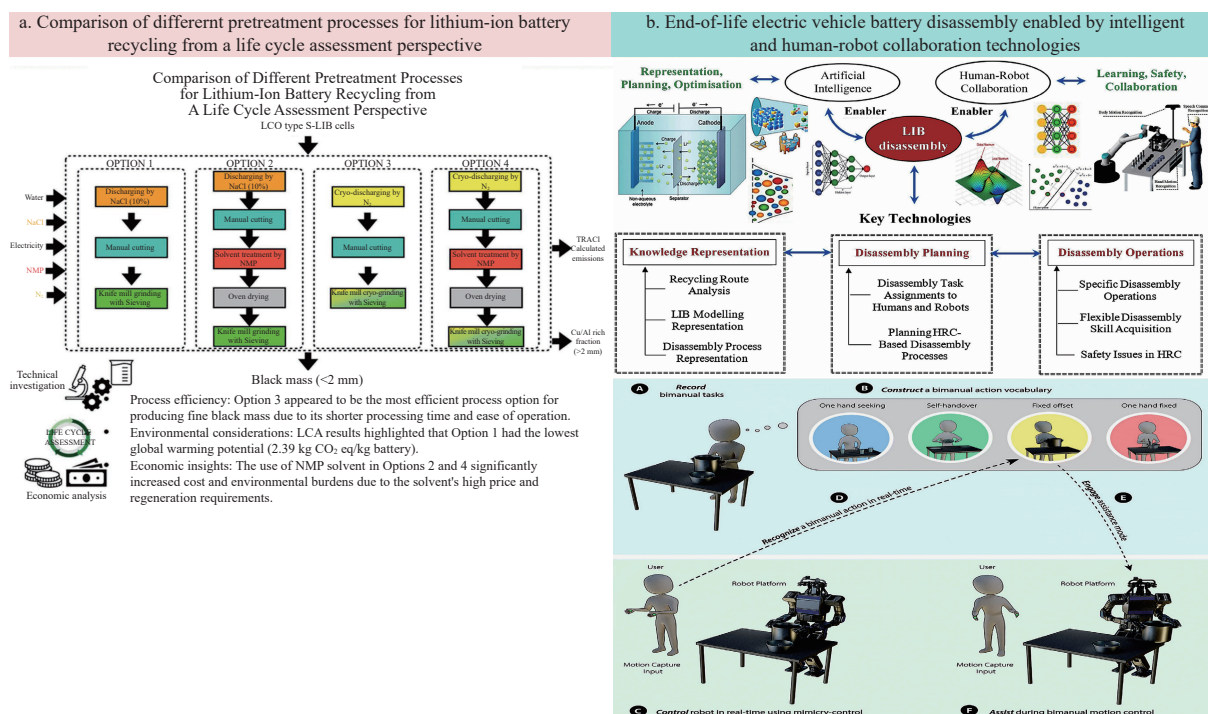


Fig. 5. Pretreatment process for spent lithium-ion battery recycling. (a) Reproduced with permission from Ref. [47]. (b) Reproduced with permission from Ref. [52].

sembly, covering knowledge representation, task planning, and execution, while noting that most implementations remain confined to laboratory settings due to poor model generalization and high computational overhead. In contrast, Yuan et al.[53] situate HRC within the Industry 5.0 framework, emphasizing human-centric manufacturing and identifying gaps in dynamic adaptability and human-factor evaluation. While both studies converge on the need for digital twin integration and reinforcement learning, Li et al. prioritize algorithmic robustness, whereas Yuan et al. highlight multi-spatiotemporal modeling and self-supervised perception. Together, these complementary perspectives suggest that future HRC systems must balance computational intelligence with human-centric flexibility, while also highlighting significant challenges, including poor model reusability, weak adaptability to dynamic conditions, and a lack of human-factor evaluation. To advance the field, the authors propose that future research should prioritize the development of multi-spatiotemporal dynamic modeling, self-supervised perception systems, optimization leveraging large AI models, and deeper integration with digital twin technologies. These directions are crucial for fostering the in-depth application of HRC in green remanufacturing and the circular economy.

While current pretreatment technologies for SLIBs have established the fundamental “discharge-disassembly-crushing-sorting” framework, scale-up remains the principal challenge. When scaled to continuous production lines, laboratory-stage techniques such as pyrolysis, cryogenic freezing, flotation, and robotic disassembly are plagued by a series of engineering issues, including high energy consumption, significant emissions of corrosive gases, refrigerant accumulation, and poor thermal uniformity within modules. Current research on pretreatment technologies for SLIBs predominantly focuses on ternary systems, while lacking tailored

parameters for other chemical systems such as LFP and solid-state batteries. Inadequate impurity control and insufficient online monitoring methods will lead to additional consumption of leaching agents in subsequent processes, narrowing the process window, and ultimately hindering the establishment of closed-loop management.

Future trends will revolve around “continuous operation, adaptiveness, and near-zero emission”: developing integrated equipment that consolidates pyrolysis, cryogenic freezing, crushing, and sorting into a single continuous production line, while utilizing digital twins for real-time regulation of key variables such as temperature, atmosphere, and crushing intensity; establishing a process parameter database tailored for diverse chemical systems, coupled with AI visual recognition to dynamically optimize disassembly pathways; incorporating technologies like catalytic reforming, amine solution circulation, and micro-nano bubble flotation to achieve in-situ recovery of refrigerants and volatile fluorine-phosphorus compounds, thereby enhancing the direct remanufacturing value of by-products such as graphite and aluminum foil; centering on robotic disassembly to accomplish high-precision classification at the source, supplying subsequent acid-free leaching and closed-loop regeneration processes with high-grade, low-hazard raw materials, and thereby advancing the pretreatment stage towards a low-cost, low-emission, and high-value-upgrading transformation.

3.2. Cascaded utilization technologies for SLIBs

Cascaded utilization represents one of the important approaches for managing SLIBs[54]. Its core principle involves batteries that, although no longer capable of meeting the performance requirements of their original equipment, still possess a certain residual energy and use value. These batteries are subsequently redeployed into other application fields

through a series of technical processing and management measures^[55,56]. Cascaded utilization and disassembly recycling of SLIBs complement each other, collectively constituting an effective strategy for the management of SLIBs^[57]. During the cascaded use of SLIBs, their application in energy storage systems can significantly extend battery service life and reduce energy storage costs while effectively enhancing power quality and power supply reliability^[58]. Beyond energy storage systems, these batteries can also play significant roles in multiple fields such as telecommunication base stations, solar power plants, and household energy storage systems^[59]. The specific application scenarios are illustrated in Fig. 6.

Luo et al.^[60] established an assessment framework to analyze the spatiotemporal distribution of retired electric vehicle batteries in China and proposed a three-phase pathway of “in-vehicle use → cascaded use → remanufacturing”. The study quantified the potential of cascaded energy storage and recycling revenues. However, the model lacks experimental validation, and the reliability of its conclusions requires further verification. Zeng^[61] systematically reviewed the full-chain technology for retired power batteries encompassing “collection → testing → regrouping → energy storage cascaded utilization,” and demonstrated its economic viability in energy storage scenarios through algorithm comparisons and case studies. Wang et al.^[62] evaluated the impact of the “cascaded utilization prior to recycling” pathway for batteries on China’s critical metal security through dynamic modeling. The study revealed that the cascaded utilization rate could be elevated to 76%, substituting 40% of the metal demand for energy storage. However, this approach would delay metal recycling, resulting in peak demands for primary lithium, cobalt, and nickel in 2040 reaching 15, 56, and 6 times the domestic annual production, respectively. The research advocates for synergistic policy and technological interventions to ensure sustainable metal supply. Lv et al.^[63] established rapid sorting and precise evaluation methods for retired LFP batteries, confirming their potential for energy storage applications. The study elucidated the performance degradation mechanisms under low-temperature and high-current conditions, providing critical foundations for establishing relevant technical specifications.

The current core bottlenecks in cascaded utilization research primarily center on three critical contradictions: consistency, safety, and economic viability. Firstly, retired batteries exhibit highly heterogeneous distributions in key parameters such as capacity and internal resistance, while existing static sorting and passive balancing strategies fail to curb dynamic divergence after grouping, leading to significant degradation of system lifespan. Secondly, the thermal runaway mechanisms of batteries in low state-of-health conditions remain incompletely elucidated, and energy storage systems continue to apply safety thresholds designed for new batteries, lacking derated thermal management protocols and fire safety standards specifically tailored for retired batteries. Thirdly, laboratory-level cycle validation demonstrates substantial discrepancies from engineering applications, while secondary costs, metal price fluctuations, and regulatory gaps dis-

tort economic models, thereby inhibiting large-scale implementation.

Future research will shift toward synergistic innovation in “digital twin-thermal safety-economics-standardization”. Through multi-parameter fusion (electrical-thermal-mechanical-aging) and dynamic reconfiguration networks, the remaining lifespan of retired batteries will become predictable and replaceable. A thermal runaway database and early warning system for low state-of-health batteries will be established to develop dedicated safety thresholds and fire protection strategies. Accelerated aging protocols under real-world conditions and open-source data sharing mechanisms will be constructed to enhance the interpretability and verifiability of lifespan models. Real-time metal price linkage and battery banking business models will be introduced to balance the economic boundaries between cascaded utilization and direct recycling. Simultaneous advancement will be made in second-life labeling, carbon footprint tracking, and cross-border transportation mutual recognition under the International Electrotechnical Commission standard framework, ultimately providing profitable and regulated scaling pathways for global cascaded utilization.

3.3. Regeneration and recycling technologies for SLIBs

3.3.1. Hydrometallurgical techniques

Hydrometallurgical techniques have emerged as a core pathway for the efficient recovery of valuable metals from SLIBs. Their fundamental principle lies in the solid-to-liquid phase transfer of metal ions (e.g., Li, Co, Ni, Mn) from cathode active materials into solution using acidic or alkaline lixiviants, followed by steps such as extraction, precipitation, ion exchange, or electrochemical reduction to achieve high-purity separation. Based on the differences in leaching media, these systems can be categorized into four main routes: inorganic acids, organic acids, ammonia-ammonium salts, and deep eutectic solvents (DESS)^[64-66].

Inorganic acids (H_2SO_4 , HCl , HNO_3) are the most widely employed in industrial applications due to their strong acidity and cost-effectiveness. Through synergistic action with reducing agents (H_2O_2 , Na_2SO_3), leaching efficiencies for valuable metals including cobalt and lithium can be enhanced to exceed 95%. However, these processes are accompanied by significant challenges including the generation of hazardous gases such as Cl_2 and NO_x , along with pronounced equipment corrosion issues. Wei et al.^[67] developed a hydrometallurgical process for recovering nickel and manganese from spent sodium-ion batteries. However, this result was obtained at a laboratory scale. Whether the process maintains efficiency with industrially relevant feedstocks remains unvalidated. This process, comprising acid leaching, selective extraction, and precipitation steps, successfully produced NiO and Mn_3O_4 with purities exceeding 99%, thereby establishing a viable route for resource cycling of sodium-ion batteries.

Organic acids (e.g., citric acid, oxalic acid, ascorbic acid) are recognized for their biodegradable nature, low corrosivity, and chelation-reduction synergy, enabling selective leaching under mild conditions with leaching efficiencies reaching

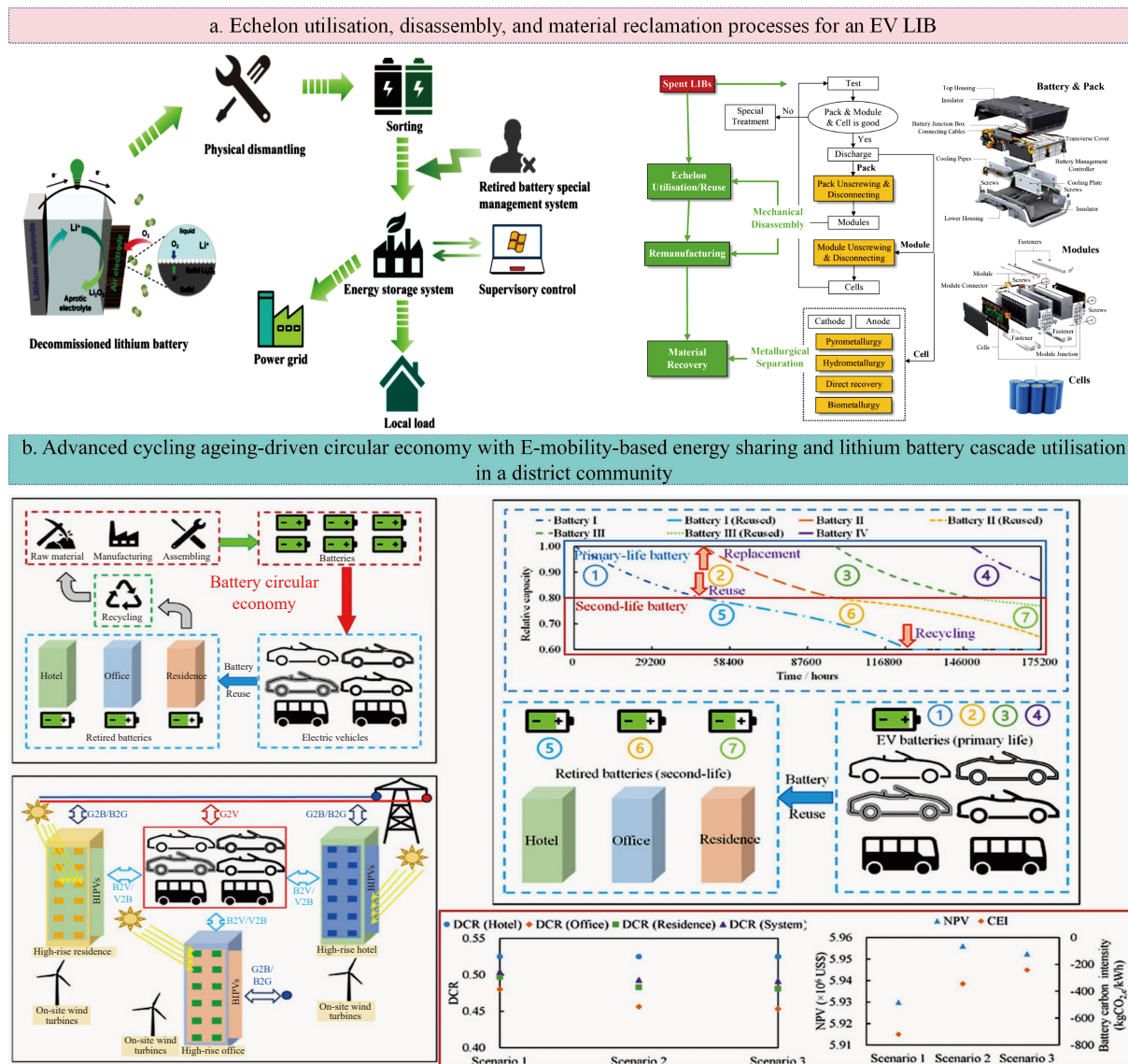


Fig. 6. Cascaded utilization of spent lithium-ion batteries. (a) Reproduced with permission from Ref. [52]. (b) Reproduced with permission from Ref. [52, 59].

90%–99%. However, their large-scale application is currently constrained by high reagent costs and prolonged reaction durations. Li et al.^[68] developed a one-step green hydrometallurgical process based on contact-electrocatalysis (CEC) for the simultaneous pretreatment and metal leaching of SLIB cathodes. The method involves directly shredding cathode sheets into fragments, which are then immersed in a malic acid solution. Under ultrasonication and with inexpensive SiO_2 as the catalyst, valuable metals are efficiently leached via the organic acid. Experimental results demonstrate that under optimized conditions (1 M malic acid, 80 mg SiO_2 , 80 °C, 6 h), leaching efficiencies of 99.6% for Li, 98.3% for Ni, 99.4% for Co, and 97.4% for Mn are achieved. This process eliminates the need for external reductants or toxic solvents, significantly simplifies conventional hydrometallurgical flow-sheets, reduces energy consumption and environmental impact, and exhibits promising potential for industrial-scale application. Zhu et al.^[69] investigated the feasibility of employing bipolar membrane electrodialysis (BMED) technology for recovering lithium hydroxide and sulfuric acid from lithium sul-

fate solutions during the hydrometallurgical recycling of SLIBs, thereby establishing a closed-loop circulation system, as schematically illustrated in Fig. 7b. The system systematically evaluated the effects of current density, membrane pair quantity, and flow rate on BMED performance, demonstrating advantages of high energy efficiency, low contamination, and favorable potential for industrial scale-up. Under operational conditions employing a 100-pair membrane stack, the theoretical energy consumption for LiOH production was reduced to 1.57 kWh kg^{-1} while achieving a Li^+ recovery rate of 99.8%. Durability tests confirmed stable membrane performance and high solution purity, with membrane fouling predominantly induced by natural aging, thus providing both technical justification and economic feasibility support for industrial implementation of BMED in lithium-ion battery (LIB) recycling. Atia et al.^[70] developed a closed-loop hydrometallurgical process for “zero-waste” recycling of SLIBs, recovering graphite, cobalt, nickel, and lithium. The process combines efficient $\text{H}_2\text{SO}_4\text{-H}_2\text{O}_2$ leaching ($\geq 98\%$ extraction of Co, Ni, Li) with integrated purification to produce Co_3O_4 (83%,

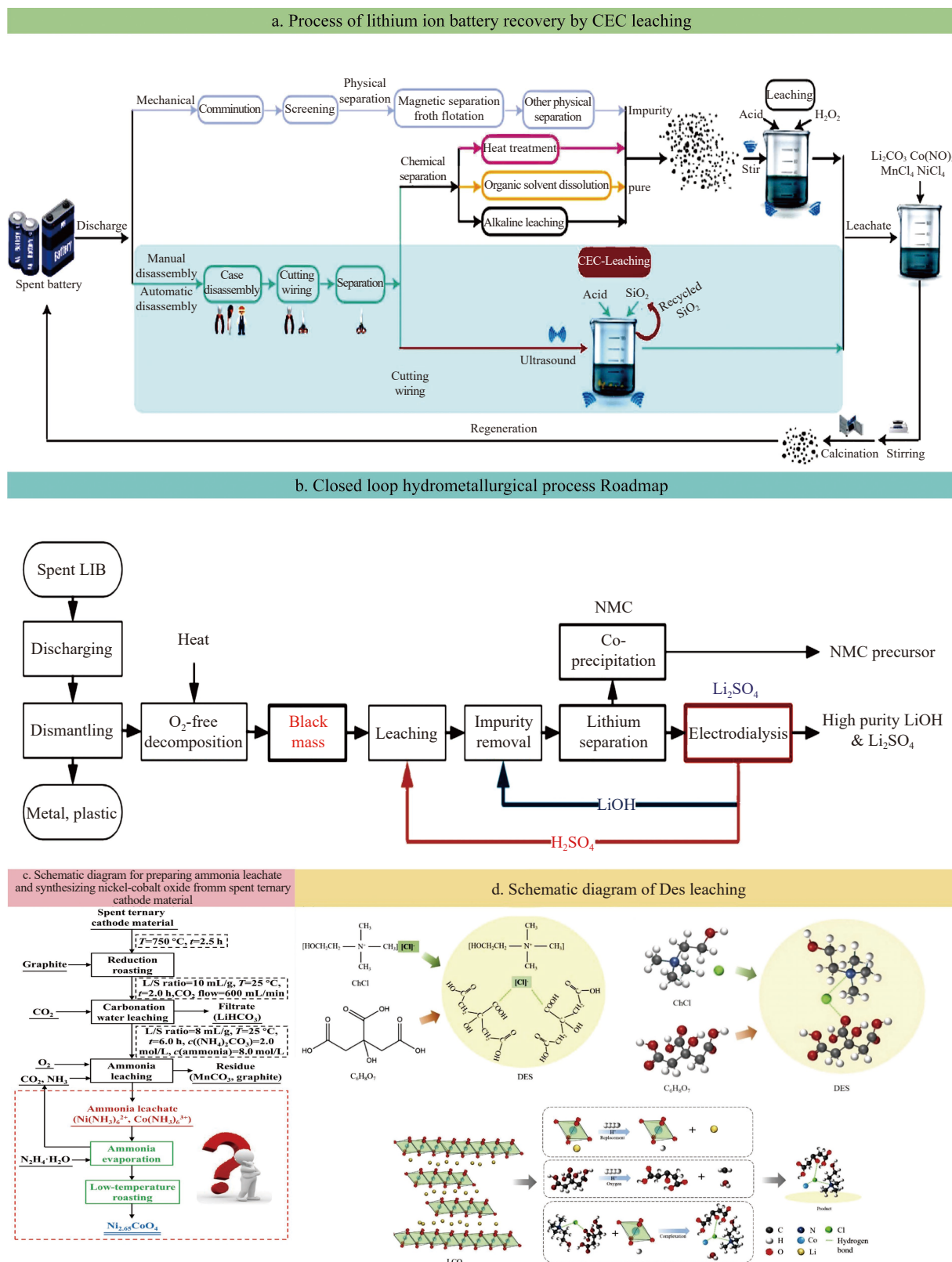


Fig. 7. Hydrometallurgical recycling strategies. (a) Reproduced with permission from Ref. [68], (b) Reproduced with permission from Ref. [69], (c) Reproduced with permission from Ref. [72], (d) Reproduced with permission from Ref. [75].

pure), CoC_2O_4 (96%), NiO (89%), Li_2CO_3 (99.8%), Na_2SO_4 (96%), and Mn-Co ferrite nanoadsorbents. Crucially, organic acids (e.g., malic and malonic acid) were shown to effectively replace H_2SO_4 , achieving comparable leaching efficiency while avoiding downstream ion interference and simplifying

wastewater treatment. The recovered Co_3O_4 and NiO were further processed into carbon-composite conversion anodes, which delivered 190 mAh g^{-1} and $\sim 180 \text{ Wh kg}^{-1}$ in full cells with lithium nickel cobalt aluminum oxide (NCA, $\text{LiNi}_x\text{Co}_y\text{Al}_2\text{O}_2$) cathodes. This “organic-leaching-to-remanufacture”

turing" route demonstrates both environmental sustainability and electrochemical viability, offering a low-acid, near-zero-waste strategy for battery recycling.

Ammonium-ammonia leaching systems (utilizing NH_3 , $(\text{NH}_4)_2\text{CO}_3$, $(\text{NH}_4)_2\text{SO}_4$, or NH_4Cl) leverage the capacity of NH_3 to form stable ammine complexes ($[\text{M}(\text{NH}_3)_n]^{2+}$) with Ni^{2+} , Co^{2+} , and Li^+ , enabling selective leaching under conditions of $\text{pH} = 9$ – 10 , temperatures of 60 – 80 °C, and with the assistance of reducing agents (Na_2SO_3 , Fe/Fe^{2+}). In contrast, Su et al.^[71] proposed a coupled process of "reduction roasting-water leaching-ammonia leaching" utilizing waste betel nut powder as a reductant for recovering valuable metals from spent ternary batteries. This process achieved preferential lithium extraction and leaching efficiencies exceeding 99% for nickel, cobalt, and manganese, while simultaneously elucidating the underlying "core-shell oxidation and layer-by-layer exfoliation" reaction mechanism. However, the lithium recovery remains appreciably lower than that achieved by conventional inorganic acid leaching, suggesting that the use of a biomass reductant introduces a trade-off between green chemistry credentials and complete lithium extraction. Yang et al.^[72] proposed a novel short-path route for recovering nickel and cobalt from the ammoniacal leachate of spent ternary LIBs with simultaneous value-added application, as illustrated in Fig. 7c. Using hydrazine as a reductant under conditions of 130 °C and 70 vol% evaporation, co-precipitation achieved recovery rates of $> 95\%$ for Ni and $> 99\%$ for Co. The resulting precursor was calcined at 300 °C for 3 h, directly synthesizing high-specific surface area mesoporous nickel-cobalt oxide $\text{Ni}_{2.65}\text{CoO}_4$. When employed as an oxygen evolution reaction electrode, the material demonstrated an overpotential of merely 350.7 mV at 10 mA cm^{-2} with a Tafel slope of $159.8 \text{ mV dec}^{-1}$, outperforming conventional NiO , Co_3O_4 , and noble metal catalysts. This strategy integrates environmentally benign recycling featuring ammonia circulation and zero discharge of high-salinity wastewater with functional material applications. A critical observation is that while the value-added application (electrode for oxygen evolution reaction) demonstrates impressive electrochemical performance, this pathway consumes the recovered nickel and cobalt rather than returning them to the battery supply chain—a deliberate choice that prioritizes immediate value generation over closed-loop battery material cycling. Ou et al.^[73] developed a "controlled-oxygen ammonia leaching-spontaneous precipitation" short-flow process for efficient and selective recovery of manganese from the black mass of spent ternary batteries. This approach achieved 96% manganese leaching under mild conditions, while enabling reagent-free spontaneous precipitation to obtain high-purity MnCO_3 with a 98% recovery rate, demonstrating advantages of process brevity, low cost, and environmental compatibility. Notably, the process selectively recovers manganese but does not address the recovery of lithium, nickel, or cobalt from the same black mass feedstock. Consequently, this method would need to be integrated with complementary processes to achieve complete resource recovery from spent ternary batteries—a system-level consideration that the study does not explore. Additionally, Li et al.^[74] created an

integrated short-flow process of "carbothermal reduction-selective ammonia leaching-sol-gel" for direct regeneration of spent nickel manganese cobalt oxide (NMC, $\text{LiNi}_x\text{Mn}_y\text{Co}_2\text{O}_2$) cathodes into high-performance lithium-rich cathode materials. The process achieved near-total leaching efficiencies of 99.3% for lithium, nickel, and cobalt with precise separation, and the regenerated cathode material exhibited exceptional electrochemical performance (initial discharge capacity of 246.5 mAh g^{-1} , energy density of 924 Wh kg^{-1}), thereby establishing a green and high-value closed-loop recycling pathway.

DESs are emerging as novel green media possessing dual functionalities of hydrogen bond complexation and reduction, enabling efficient leaching of cobalt and lithium at $> 99\%$ efficiency under mild conditions (80 – 120 °C, atmospheric pressure) without the need for external acids/reducing agents. These solvents exhibit advantages including ready biodegradability and recyclability. However, their industrial implementation faces challenges such as prolonged leaching durations (48 – 72 h) and mass transfer limitations caused by high viscosity. Niu et al.^[75] employed a choline chloride-citric acid DES as a green leaching system. As illustrated in Fig. 7d, under mild conditions of 80 °C, 60 min, solid-to-liquid ratio of 20 g L^{-1} , and molar ratio of 2:1, efficient leaching of cobalt (94.26%) and lithium (96.98%) from spent lithium cobalt oxide cathode materials was achieved. Kinetic and spectroscopic analyses confirmed a mechanism involving preferential lithium delithiation, followed by citric acid-mediated reduction of Co^{3+} and subsequent dissolution via chloride complexation. Cobalt was subsequently precipitated as oxalate with a recovery rate of 94.86%, while lithium was recovered via carbonate precipitation, thereby establishing a closed-loop recycling pathway for both metals. Zou et al.^[76] constructed a ternary DES comprising choline chloride, ethylene glycol, and maleic acid, which achieved efficient leaching of multiple metals (Ni, Co, Mn, Li, Fe, Cu, all with leaching efficiencies $> 99\%$) from SLIB black mass under conditions of 80 °C, 2 h, and a liquid-to-solid ratio of 50 mL g^{-1} . Through sequential chemical precipitation and electrodeposition methods, gradual separation and recovery of individual metals were accomplished, ultimately yielding high-purity graphite, Fe precipitates, Cu metal, Ni-Co alloy, $\text{MnO}(\text{OH})$, and Li_2CO_3 products. The study systematically investigated the leaching mechanisms of the DES and metal separation processes, providing a novel strategy for the green recovery of complex mixed materials. Xu et al.^[77] developed a ternary DES system that achieved highly efficient leaching of lithium, nickel, cobalt, and manganese from spent NMC cathodes under mild conditions, with leaching efficiencies all approaching 99%. Subsequently, through step-wise separation and precipitation procedures, four high-purity metal products were successfully recovered, demonstrating overall recovery rates all exceeding 86%. This work thereby establishes a novel pathway for green and energy-efficient recycling of lithium battery cathode materials. Wen et al.^[78] utilized a choline chloride-malononic acid-2-pyrrolidinone DES to simultaneously achieve delamination of spent LiCoO_2 cathodes from aluminum foil (exceeding 98%) and metal leaching (Li 97.6%, Co 96.1%, Al $< 4\%$) in a single step.

The DES disrupts the C–F bonds of the PVDF binder while simultaneously reducing Co^{3+} to Co^{2+} to form $[\text{CoCl}_4]^{2-}$ complexes, thereby creating a mutual promotion of delamination and leaching. Recovery of $\text{CoC}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$ and Li_2CO_3 was accomplished via oxalate and carbonate precipitation, with the DES maintaining leaching efficiencies above 90% even after four recycling cycles. This strategy, operating under mild conditions (100 °C, 30 min), simplifies the process flow and reduces reagent consumption, providing a novel pathway for green and efficient recycling of LIBs.

Several critical bottlenecks persist in the hydrometallurgical recycling of SLIBs. In the leaching stage, inorganic acids are confronted with environmental pollution and equipment corrosion, organic acids exhibit high costs and low efficiency, while DESs face challenges in scaling up due to their high viscosity and prolonged leaching cycles. Regarding the separation stage, the removal of impurity elements remains difficult, and multi-metal stepwise recovery processes are complex with substantial wastewater generation. Overall, the technology is constrained by insufficient scalability and high costs, which hinder its widespread adoption.

Future technological development should concentrate on: developing efficient and environmentally benign novel leaching systems, optimizing the performance of DESs, and exploring synergistic leaching processes; researching highly selective and cost-effective separation materials and methods to enhance metal recovery efficiency and product purity; and strengthening engineering scale-up and integrating green processes to promote reagent recycling and emission reduction, ultimately achieving sustainable and large-scale recycling.

3.3.2. Pyrometallurgical techniques

The principle of pyrometallurgical recycling centers on “high-temperature treatment,” achieving separation of valuable metals from impurities through various processes^[79]. Among

these, high-temperature smelting directly utilizes elevated temperatures to melt metallic components within batteries, thereby achieving separation and recovery. Reduction roasting involves the introduction of reducing agents under high-temperature conditions to reduce metal oxides into metallic forms, facilitating subsequent recovery. Salt-assisted roasting employs salt compounds as additives to promote the decomposition or modification of battery materials during the roasting process, thereby enhancing metal recovery rates^[80].

Zhu et al.^[81] proposed a co-smelting process for waste WPCBs and copper slag (Fig. 8a): debromination at 500 °C, then co-smelting at 1300 °C under 30% O_2 , with slag as flux and pyrolytic carbon as reductant, achieving one-step recovery of Cu–Pb–Sn alloy with rates of 95.6%, 82.5%, and 68.8%. The reaction mechanisms were clarified, offering a feasible pyrometallurgical route for solid waste synergy. Wang et al.^[82] proposed an innovative “co-roasting-magnetic separation” process integrating spent NMC523 cathodes with silicon carbide waste (SCW), as illustrated in Fig. 8b. Utilizing Al in SCW as the reductant and residual carbon as an auxiliary under conditions of 700 °C for 60 min, the process reduces Ni/Co in the cathode to metallic forms while converting Mn to MnO and combining Li–Al into LiAlO_2 . Following 3-minute grinding and magnetic separation at 290 kA m^{-1} , a magnetic concentrate containing 58.7% Ni and 21.5% Co with recovery rates exceeding 70% is obtained, along with non-magnetic tailings enriched with Li–Al–Si–Mn. This strategy thereby accomplishes low-carbon, efficient, and selective recovery of two hazardous wastes, while elucidating the mechanisms of Al–C synergistic reduction and magnetic separation. Feng et al.^[83] proposed a “spatially separated co-roasting” strategy, as illustrated in Fig. 8c, where SLIB cathode powder and waste biomass are placed in separate upper and lower layers. Utilizing biomass pyrolysis gases (primarily CO) under conditions of 650 °C for 20 min at a mass ratio of 1:2, the cathode materi-

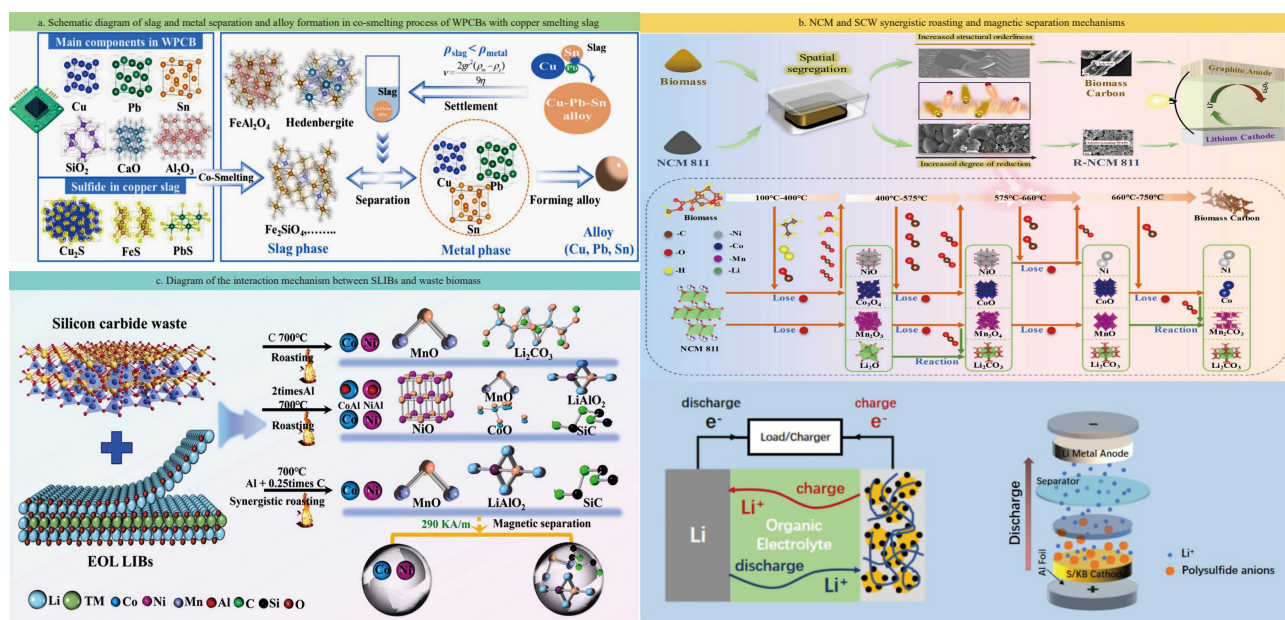


Fig. 8. Pyrometallurgical recycling strategies. (a) Reproduced with permission from Ref. [81], (b) Reproduced with permission from Ref. [82], (c) Reproduced with permission from Ref. [83].

als are reduced, achieving a selective lithium leaching efficiency of 99.6% via water extraction, while the released O^{2-} anions promote biomass carbon graphitization, yielding supercapacitor carbon with a specific capacitance of 125 F g^{-1} . This process demonstrates applicability to various cathode materials and biomass types, generating a profit of 2790 USD per ton of spent batteries, outperforming conventional hydrometallurgical/pyrometallurgical processes in simplicity, efficiency, and energy consumption, thereby enabling synergistic valorization of dual wastes. Addressing the emergence of silicon-based anode batteries, Chen et al.^[84] developed a “laser-silicon thermic” low-carbon recycling process, employing silicon as a reductant under laser irradiation to rapidly reduce LiCoO_2 into a Co-Si alloy with simultaneous slag formation. This method achieves significant weight reduction with CO_2 emissions reduced to merely half of conventional carbothermal reduction, providing a novel solution for clean recycling.

Pyrometallurgical recycling of SLIBs confronts multiple challenges in practical applications: high energy consumption and elevated costs, driven by high-temperature processing and substantial equipment investments; low metal recovery rates, particularly for lithium where recovery often falls below 50%—a limitation stemming from high-temperature lithium volatilization and slag entrainment, as summarized in Table 1; significant environmental pollution risks, including potential release of toxic gases such as hydrogen fluoride and secondary pollution from salt-assisted roasting; and notable equipment corrosion and safety hazards, encompassing HF-induced equipment degradation and potential combustion/explosion risks from undischarged batteries. Additionally, existing technologies exhibit limited adaptability to emerging battery systems such as high-nickel, high-voltage, and solid-state batteries.

To break through current bottlenecks, the future strategic focus on pyrometallurgical recycling technologies should prioritize the following directions: First, promoting green and low-carbon transition by optimizing heat recovery systems and integrating renewable energy sources such as solar power to reduce energy consumption and carbon emissions, thereby contributing to the carbon peaking and carbon neutrality goals. Second, enhancing multi-technology integration by coupling pyrometallurgy with hydrometallurgical and physical separation techniques to establish diversified recycling systems that improve metal recovery rates and product purity. Third, accelerating intelligent and digital upgrades

through the introduction of AI and big data technologies for real-time monitoring and optimization of process parameters, thereby enhancing production efficiency and mitigating operational risks. Fourth, deepening fundamental research on reaction mechanisms, with focused investigation on metal redox processes and slag formation mechanisms, to provide theoretical support for process innovation. Fifth, improving adaptability to next-generation batteries by developing specialized pyrometallurgical recycling strategies tailored to high-nickel, high-voltage, and solid-state battery systems, ensuring efficient circular utilization of future battery materials.

Table 1 reveals that lithium recovery in pyrometallurgical processes is highly sensitive to operating temperature, atmosphere, and slag chemistry. The generally low recovery (< 50% in conventional routes) primarily results from high-temperature volatilization and irreversible incorporation into silicate/aluminate slag phases. Vacuum-based and salt-assisted roasting approaches show improved lithium recovery by suppressing volatilization or enabling subsequent water leaching, albeit with added process complexity.

3.3.3. Bioleaching techniques

The core of bioleaching technology involves the utilization of microorganisms and their metabolites to achieve the dissolution and transfer of valuable metals from SLIBs^[94]. Its fundamental principle relies on microorganisms as the central driving force, whose metabolic activities (e.g., acid production and redox reactions) create the necessary conditions for metal leaching. During this process, target metals within the batteries are transformed from solid compounds into ionic species in solution, entering the leachate and thereby accomplishing the solid-to-liquid transfer^[95]. Bioleaching technologies can be primarily categorized into three types: The first is bioacidolysis leaching, which utilizes organic acids (e.g., citric acid, oxalic acid, and gluconic acid) produced by microbial metabolism to dissolve metals, disrupting the structure of metal compounds either through direct acid attack or via complex formation. The second involves redox reactions, which rely on the redox capabilities of autotrophic bacteria to alter metal valence states. The key mechanism here is electron transfer between microorganisms and metal compounds, thereby converting insoluble compounds into soluble forms. The third category comprises complexation reactions, where chelating agents or organic salts produced by microbes form stable complexes with metal ions, thereby enhancing metal solubility and stability in solution and facilitating their release

Table 1. Distribution of lithium losses across representative pyrometallurgical processes for SLIB recycling^[85–93].

Process type	Temperature (°C)	Atmosphere	Lithium recovery (%)	Lithium distribution/loss mechanism
Carbothermal reduction roasting	650–850	Inert (Ar/N ₂)	35–55	Li volatilized as Li(g) or LiOH(g); 20%–40% retained in slag phase
Salt-assisted roasting (Na ₂ SO ₄)	800–950	Air	60–75	Li converted to water-soluble Li ₂ SO ₄ ; losses via slag entrainment (10%–25%)
Conventional smelting (reductive)	1200–1450	Reducing (CO)	20–40	Severe Li volatilization (> 60%) as Li ₂ O(g); minor Li in alloy phase
Vacuum distillation	1000–1200	Vacuum (< 100 Pa)	65–85	Enhanced Li recovery via vapor condensation; residue mainly Co, Ni, Mn
Co-smelting with slag formers	1300–1500	Oxidizing	< 30	Most Li (> 70%) incorporated into slag (Li ₂ SiO ₄ , LiAlO ₂), unrecoverable

from the solid phase^[96].

Yun et al.^[97] developed a process for the effective separation of impurities (Al, Cu, Fe) and recovery of valuable metals (e.g., Co) from iron-rich bioleaching solutions. As illustrated in Fig. 9a, solvent extraction was employed using 20% D2EHPA at pH 4.0 to extract Fe^{3+} , Al^{3+} , Cu^{2+} , and Mn^{2+} , followed by two-stage washing with MnSO_4 solution to recover co-extracted Co^{2+} . Finally, concentrated sulfuric acid and aqua regia were utilized for stripping the impurity metals. The study validated the feasibility of this approach for efficient iron removal and selective cobalt recovery, providing new insights for treating bioleachates from SLIBs. Roy et al.^[98] investigated the feasibility of employing *Acidithiobacillus ferrooxidans* (NCIM 5370) for efficient bioleaching of critical metals including lithium, cobalt, manganese, and nickel from mixed cathode materials of SLIBs. As illustrated in Fig. 9b, the study systematically evaluated the impact of varying concentrations of Fe^{2+} ($1\text{--}7\text{ g L}^{-1}$) and elemental sulfur S^0 ($0.5\text{--}5\text{ g L}^{-1}$) as energy sources on bioleaching efficiency. The combined use of $3.5\text{ g L}^{-1}\text{ S}^0$ and $5\text{ g L}^{-1}\text{ Fe}^{2+}$ was found to achieve extraction efficiencies of 100% for both lithium and cobalt, and $>99.8\%$ for both nickel and manganese. Subsequent treatment steps involving chemical precipitation and solvent extraction enabled highly selective recovery of the target metals (Li: 96%, Co: 98%, Mn: 89%, Ni: 82.4%) with purities all exceeding 95%. Furthermore, techno-economic analysis demonstrated the economic viability of the process, with a net profit of \$0.12 per unit of cathode active material (CAM) processed and an investment payback period of 2.6 years, highlighting its potential for industrial-scale application. Alipanah et al.^[99] conducted a comprehensive study on the recovery of critical metals (lithium, cobalt, nickel, manganese) from SLIBs via gluconic acid-based bioleaching, proposing an economic optimization strategy aimed at maximizing net present value (NPV). Integrating thermodynamic modeling with response surface methodology, the study systematically optimized key parameters including temperature, pulp density, time, and gluconic acid concentration. Under the identified optimal conditions (55°C , 2.5% pulp density, 30 h, 75 mM gluconic acid), recovery rates reached 86% for cobalt, 84% for nickel, and 100% for both lithium and manganese, with an associated NPV of \$92 million, thereby demonstrating significant applica-

tion potential. Gu et al.^[100] employed an acidophilic microbial consortium for bioleaching to recover lithium from spent LFP batteries. Through process optimization, a lithium leaching concentration of 1.6 g L^{-1} was achieved at a high pulp density of 125 g L^{-1} , and Li_3PO_4 was subsequently obtained via precipitation with a lithium recovery rate of 89%. The study confirmed that this process exhibits a lower carbon footprint compared to conventional methods, thus establishing it as a sustainable recycling approach. Xia et al.^[101] pioneered the use of food-grade *Lactobacillus acidophilus* for highly selective bioleaching of lithium from spent LFP batteries. What distinguishes this study is the explicit comparison: lithium leaching reached 77.1% while iron leaching was only 5.8%, a selectivity factor that substantially exceeds that of conventional acid leaching, demonstrating that metabolic pathway engineering can address the selectivity challenge inherent to chemical methods. In a two-step bioleaching process, a lithium leaching efficiency of 85.1% was achieved. Furthermore, its extracellular polymeric substances (EPS) significantly enhanced the leaching selectivity (lithium: 77.1% vs. iron: 5.8%). The study revealed that lactic acid served as the primary leaching driver, while EPS functioned through enhanced adsorption and toxicity resistance mechanisms, thereby providing a novel strategy for electronic waste recycling. Shokri et al.^[102] developed a green leaching process (SCMB) utilizing *Acidithiobacillus* species to generate bio-sulfuric acid (BSA) for lithium recovery from spent nickel manganese cobalt oxide (NMC, $\text{LiNi}_x\text{Mn}_y\text{Co}_z\text{O}_2$) batteries. This process achieved a lithium recovery rate of 95.2% through two cycles of high-pulp-density (100 g L^{-1}) leaching without requiring chemical reducing agents, subsequently yielding Li_2CO_3 with a purity exceeding 99%. Life cycle assessment confirmed that the environmental impact of this technology is substantially lower than conventional hydrometallurgical methods, establishing it as a sustainable recycling pathway.

Currently, bioleaching technology faces significant bottlenecks in industrial-scale applications: low leaching efficiency and prolonged operational durations; limited tolerance of microorganisms to metal ions; stringent control requirements for process parameters; a complex battery composition adversely affecting mass transfer efficiency; and an incomplete framework for comprehensive environmental risk

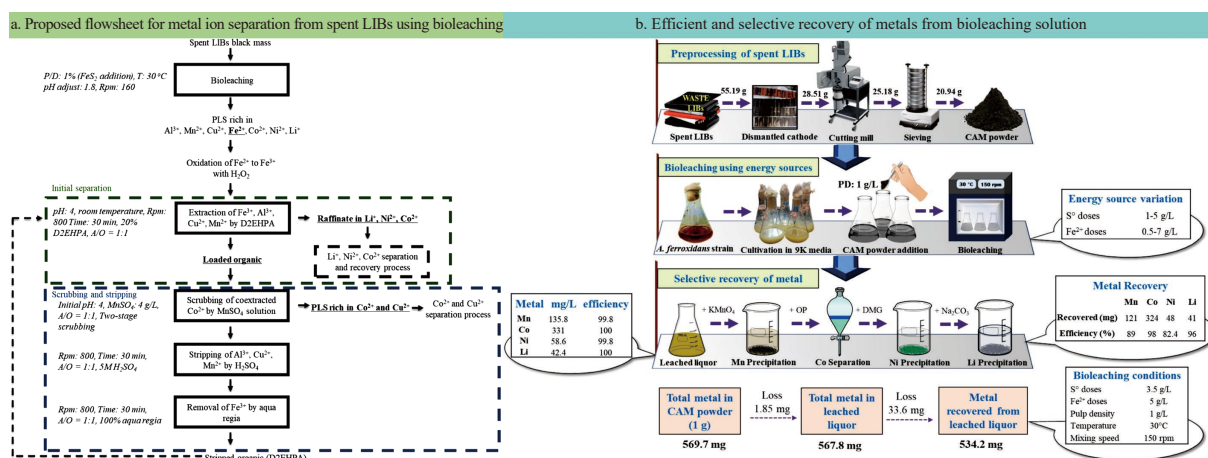


Fig. 9. Bioleaching recycling strategies. (a) Reproduced with permission from Ref. [97], (b) Reproduced with permission from Ref. [98].

assessment.

To address the aforementioned challenges, future development should prioritize five strategic directions: enhancing process efficiency through systematic optimization and response surface methodology; developing robust microbial strains with heightened tolerance and activity via strain selection or genetic engineering; establishing standardized lifecycle environmental risk assessment protocols; exploring synergistic integration of bioleaching with hydrometallurgical and electrochemical technologies; and further elucidating microbial-metal interaction mechanisms to provide theoretical foundations for technological breakthroughs.

3.4. Comparative analysis of mainstream recycling technologies

The three mainstream recycling technologies for valuable resources from SLIBs each demonstrate distinct characteristics, as summarized in Table 2. Hydrometallurgical techniques achieve the highest metal recovery rates, demonstrating particular advantages in lithium recovery^[103,104], though they are characterized by high capital investment and energy consumption, along with potential wastewater pollution risks^[105]. Pyrometallurgical methods offer large processing capacity and relatively lower operational costs^[106]. However, substantial lithium losses and significant carbon emissions represent their primary limitations^[107,108]. Bioleaching technologies are recognized as the most environmentally benign and cost-effective approach^[109], but the extended processing dura-

tion constitutes the critical bottleneck for industrial implementation^[110]. In summary, hydrometallurgy leads in metal recovery efficiency, pyrometallurgy excels in processing throughput, while bioleaching demonstrates superior environmental compatibility. Consequently, the selection of an appropriate technological pathway requires comprehensive consideration of target battery chemistry, prioritized value recovery objectives, and specific environmental requirements.

Therefore, the selection of technological pathways must be based on a comprehensive trade-off considering the target battery types, priority value recovery objectives, and environmental requirements. Looking forward, the development of these three technological pathways should focus on leveraging their respective strengths, mitigating weaknesses, and pursuing synergistic integration. For hydrometallurgy, the R&D focus should prioritize process intensification, reagent recycling, and the development of green solvents to significantly reduce its energy consumption and environmental footprint. For pyrometallurgy, future development should be directed towards upgrading energy recovery systems and achieving technological breakthroughs in lithium enrichment and recovery, fundamentally addressing its inherent drawbacks of high carbon emissions and significant lithium losses. Meanwhile, for bioleaching, the central mission requires dedicated efforts to cultivate high-efficiency microbial strains via genetic engineering and optimize the reaction environment, aiming at drastically shortening the processing duration and accelerating its industrial adoption. Furthermore, exploring hybrid pro-

Table 2. Comparative analysis of core indicators for three mainstream recycling technologies of spent lithium-ion battery resources^[103–112].

Core indicator	Hydrometallurgy	Pyrometallurgy	Bioleaching
Basic principle	Dissolution of metal ions in acid/alkaline media, followed by separation and purification via extraction, precipitation, etc.	High-temperature (600–1300 °C) smelting/reduction for metal recovery through metallurgical and magnetic separation processes.	Microbial metabolism produces acids or chelators to dissolve metals, relying on biological reduction/oxidation.
Metal recovery rate	High: Ni, Co: > 90%, Li: > 95%	Moderate: Co, Ni: > 80%, Li: 40%–50%	High after optimization: Co leaching up to 93%, Li leaching up to 91%
Energy consumption (kWh/kg black mass)	High: 100–200 (solution heating, agitation, separation)	Moderate: 150–300 (energy-intensive, but partial heat recovery is feasible)	Low: 10–30 (ambient temperature and pressure, mainly for agitation and aeration)
Cost (reagent cost, USD/t spent batteries)	High: 800–1500 (high reagent consumption and wastewater treatment costs)	Moderate to low: 200–500 (short process, low reagent use, but high energy costs)	Low: 50–200 (minimal reagent costs, though extended cycle time increases indirect expenses)
Processing time	Short to moderate: 3–7 d (from leaching to product recovery)	Short: 2–10 h (high-temperature core step) + subsequent treatment	Very long: 20–60 d (limited by microbial growth and reaction kinetics)
Environmental impact (CO ₂ emissions, kg CO ₂ /kg black mass)	Moderate: 50–100 (main risks: fluoride-containing wastewater and dissolved transition metals such as Co, Ni, and Mn requiring purification)	High: 150–400 (high carbon emissions, flue-gas pollution requiring purification)	Very low: 5–15 (no high-temperature harmful gases, minimal wastewater)
Equipment investment	High (corrosion-resistant reactors, extraction units, complex water-treatment systems)	Moderate (high-temperature furnaces, flue-gas treatment systems, standardized process equipment)	Low (simple reactors, no need for high-temperature/pressure or complex chemical equipment)
Applicable battery types	Relatively simple compositions, e.g., LFP, low-nickel ternary batteries	Complex, high-value-metal-content systems, e.g., high-nickel ternary batteries, highly adaptable	Low-grade spent batteries, battery scraps, etc.
Scalability assessment	High: mature industrial process, easily scalable. Challenge: large-scale water-treatment systems.	High: compatible with existing smelting infrastructure, suitable for large-scale centralized processing. Challenge: stringent environmental regulations and carbon-reduction pressure.	Low to moderate: large reactor footprint and long cycles pose significant industrial bottlenecks. Prospect: suitable for low-grade feedstocks or as an eco-friendly pretreatment option.

cesses such as “pyrometallurgical pretreatment coupled with hydrometallurgical refining” or “bioleaching followed by hydrometallurgical purification” to form integrated solutions with complementary advantages will be a crucial direction for enhancing both the economic viability and sustainability of the entire recycling industry.

4. Future development trends

4.1. Direct regeneration and repair

The core principle of direct regeneration and repair technologies lies in avoiding material-level disassembly of batteries, instead directly restoring their electrochemical performance through specific processes^[113]. This technology primarily encompasses three fundamental principles: (i) Electrochemical regeneration repairs electrode structures and eliminates polarization by adjusting electrical parameters (e.g., voltage, current) or employing specialized electrochemical techniques (e.g., pulse repair), thereby restoring battery capacity and cycle life. (ii) Solid-phase regeneration improves the crystal structure and interfacial states of electrode materials under solid-state conditions by controlling temperature, pressure, or adding healing agents, with the entire process occurring in the absence of liquid media. (iii) Hydrothermal repair utilizes high-temperature and high-pressure aqueous environments to induce reactions such as dissolution-recrystallization or surface modification of electrode materials, thereby repairing microstructures and optimizing interfacial characteristics^[114,115]. Owing to their distinct mechanisms and processes, these direct regeneration and repair technologies exhibit differentiated advantages and limitations^[116]. Electrochemical regeneration is characterized by operational simplicity, mild conditions, and ease of automation. However, its effectiveness is limited for batteries with severe structural degradation, and the performance stability post-repair may be inadequate^[117]. Solid-phase regeneration offers the advantage of avoiding secondary contamination from liquid media and features a more streamlined process flow. Conversely, it demands extremely stringent control over process parameters, involves high equipment costs, and demonstrates limited efficacy in repairing severely degraded materials^[118]. Hydrothermal repair enables deep and uniform repair of microstructural defects, achieves high-capacity recovery rates, and allows for simultaneous material modification. Notably, its drawbacks include reliance on expensive high-temperature and high-pressure equipment, high energy consumption, prolonged processing duration, and risks of corrosion or secondary damage^[119].

Zhou et al.^[120] proposed a value-added reuse strategy for LFP cathode materials, as illustrated in Fig. 10a. Through hydrothermal treatment at 220 °C for 2 h using ascorbic acid as a reductant and Li_2CO_3 as a lithium source, lattice reconstruction was achieved to regenerate LFP, which was subsequently employed as an adsorbent for selective lithium extraction from acidic leachate. This method requires no high-temperature calcination and demonstrates exceptional lithium adsorption capacity (38.5 mg g^{-1}) with remarkable cycling stability. It maintains efficient lithium recovery even at low concentrations (20 ppm) without causing electrode dissolution. Thermodynamic and electrochemical analyses revealed the

synergistic mechanism between vinylene carbonate's strong reducibility and high temperature in critical structural repair of LFP. The established closed-loop recycling pathway balances economic viability with environmental sustainability, thereby providing a novel paradigm for resource recovery from SLIBs. Zhong et al.^[121] established an ultralow-voltage (0.5 V) electrochemical leaching system utilizing metallic nickel as the anode, achieving highly efficient and impurity-free dissolution of Ni, Co, Mn, and Li (> 98 %) from spent NMC523 cathodes. The study elucidated the interfacial coupling mechanism of “anodic oxidation-cathodic reduction-dissolution”, as illustrated in Fig. 10b: continuous anodic dissolution of Ni generates Ni^{2+} and releases electrons, while external circuit electrons synergize with trace ascorbic acid to reduce high-valence metals in NMC, effectively suppressing oxygen evolution and side reactions. The resulting nickel-rich leachate required no purification and was directly upgraded to NMC622 cathodes via oxalate co-precipitation and solid-state sintering, demonstrating superior cycling performance over 100 cycles compared to pristine materials. With process energy consumption reduced to 15.89 kWh t^{-1} , this represents the electrochemical recycling route for spent LIBs with the lowest operational voltage and energy consumption reported to date. Liao et al.^[122] developed a closed-loop regeneration strategy based on potassium sodium tartrate (PST)-mediated reduction and lattice reconstruction for efficient, low-energy, high-value repair of degraded LFP cathode materials. The PST functions through a dual mechanism: its tartrate moiety acts as a reducing agent to convert Fe^{3+} (from degraded LiFePO_4) back to Fe^{2+} , while its potassium and sodium ions facilitate lithium replenishment by creating a favorable ionic environment for relithiation, thereby enabling simultaneous restoration of the olivine crystal structure. As illustrated in Fig. 10c, direct hydrothermal regeneration simultaneously achieved lithium replenishment, Fe^{3+} reduction, and lattice defect repair. The regenerated LFP (RA-LFP) exhibited a specific capacity of 143 mAh g^{-1} at 1C rate, maintained > 99% capacity retention after 150 cycles, and preserved 82.9% capacity following 1000 cycles at a high rate of 10C. This approach requires no noble metal catalysts, generates only CO_2 and H_2O as by-products, and demonstrates advantages including low cost, minimal carbon emissions, and high profitability, thereby establishing a sustainable solution for closed-loop recycling of retired LFP batteries. Yang et al.^[123] developed a streamlined process for the direct regeneration of high-voltage Mn-rich cathode materials ($0.2\text{Li}_2\text{MnO}_3 \cdot 0.8\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$) from mixed SLIB cathodes. This acid leaching-coprecipitation-solid-state-sintering route successfully yielded regenerated cathode materials with well-defined structure and uniform particle size ($D_{50} = 7.35 \text{ }\mu\text{m}$), without requiring the separation of Ni, Co, and Mn. The regenerated material exhibited excellent electrochemical performance, delivering a discharge capacity of 248.3 mAh g^{-1} at 0.1 C and retaining 167 mAh g^{-1} at 1 C. After 50 cycles, capacity retention rates reached 88%, 85%, and 80% under various cycling conditions, surpassing those of conventionally blended cathode materials. Furthermore, clean production analysis demonstrated that the process enables effective treatment of gaseous, liquid, and solid waste streams,

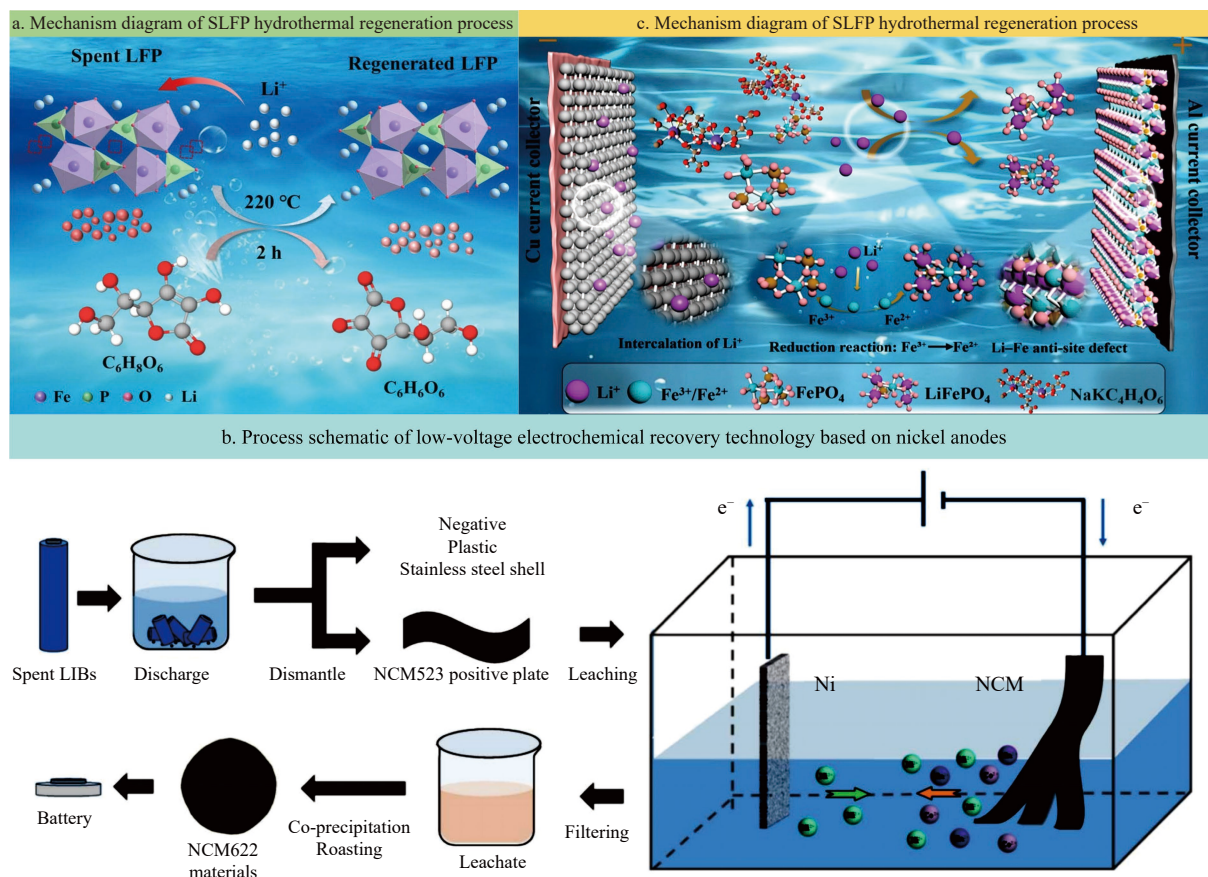


Fig. 10. Direct regeneration and repair recycling strategies. (a) Reproduced with permission from Ref. [120], (b) Reproduced with permission from Ref. [121], (c) Reproduced with permission from Ref. [122].

offering high resource utilization efficiency and a favorable environmental profile.

Current direct regeneration and repair technologies for SLIBs face several major bottlenecks: Firstly, the remediation efficacy remains limited—electrochemical regeneration struggles to effectively remediate severe structural damage and exhibits inadequate post-repair performance stability, while both solid-phase and hydrothermal repairs show constrained effectiveness on deeply degraded materials. Secondly, process control poses significant challenges, as solid-phase regeneration requires precise regulation of temperature, pressure, and healing agent dosage, thereby increasing process complexity and cost. Thirdly, equipment costs are prohibitively high—hydrothermal repair relies on expensive high-temperature/high-pressure apparatus, accompanied by substantial energy consumption, prolonged durations, and corrosion risks, which collectively hinder large-scale application. Fourthly, systematic technical solutions are lacking, particularly in the cascaded utilization sector where insufficient understanding of intrinsic safety mechanisms and internal thermochemical reactions restricts deployment in high-safety-demand scenarios. Lastly, detection efficiency remains suboptimal due to the absence of specialized lifespan assessment technologies and dedicated equipment, resulting in cumbersome procedures, elevated costs, and impediments to industrial scaling.

Future development should prioritize the following directions to achieve breakthroughs: optimizing process param-

eters through systematic investigation to enhance the stability of regeneration efficacy and reduce sensitivity to operational conditions; developing novel low-cost, high-efficiency equipment, such as improved electrochemical remediation systems and advanced high-temperature/high-pressure reactors, to mitigate application costs and risks; strengthening fundamental research to elucidate internal battery reaction mechanisms and material structure evolution, thereby providing theoretical foundations for technological innovation; creating efficient detection technologies and specialized equipment to enable rapid and accurate lifespan assessment, facilitating large-scale implementation; and exploring environmentally benign processes, such as developing novel lithium carriers and other highly effective regeneration methods, to balance ecological compatibility with cost-effectiveness, ultimately promoting the sustainable advancement of this technology.

4.2. Electrochemical recycling technologies

Electrochemical recycling technologies for SLIBs primarily achieve the separation and recovery of valuable metals through electrochemical reactions. The fundamental principle relies on two core mechanisms: first, the principle of electrochemical deposition, which utilizes electrode reactions to reduce target metal ions (e.g., cobalt, nickel) in solution, causing them to deposit as elemental metals or compounds on the cathode; second, the principle of electrodialysis, which employs ion-exchange membranes under an electric field to drive the selective migration and enrichment of specific ions

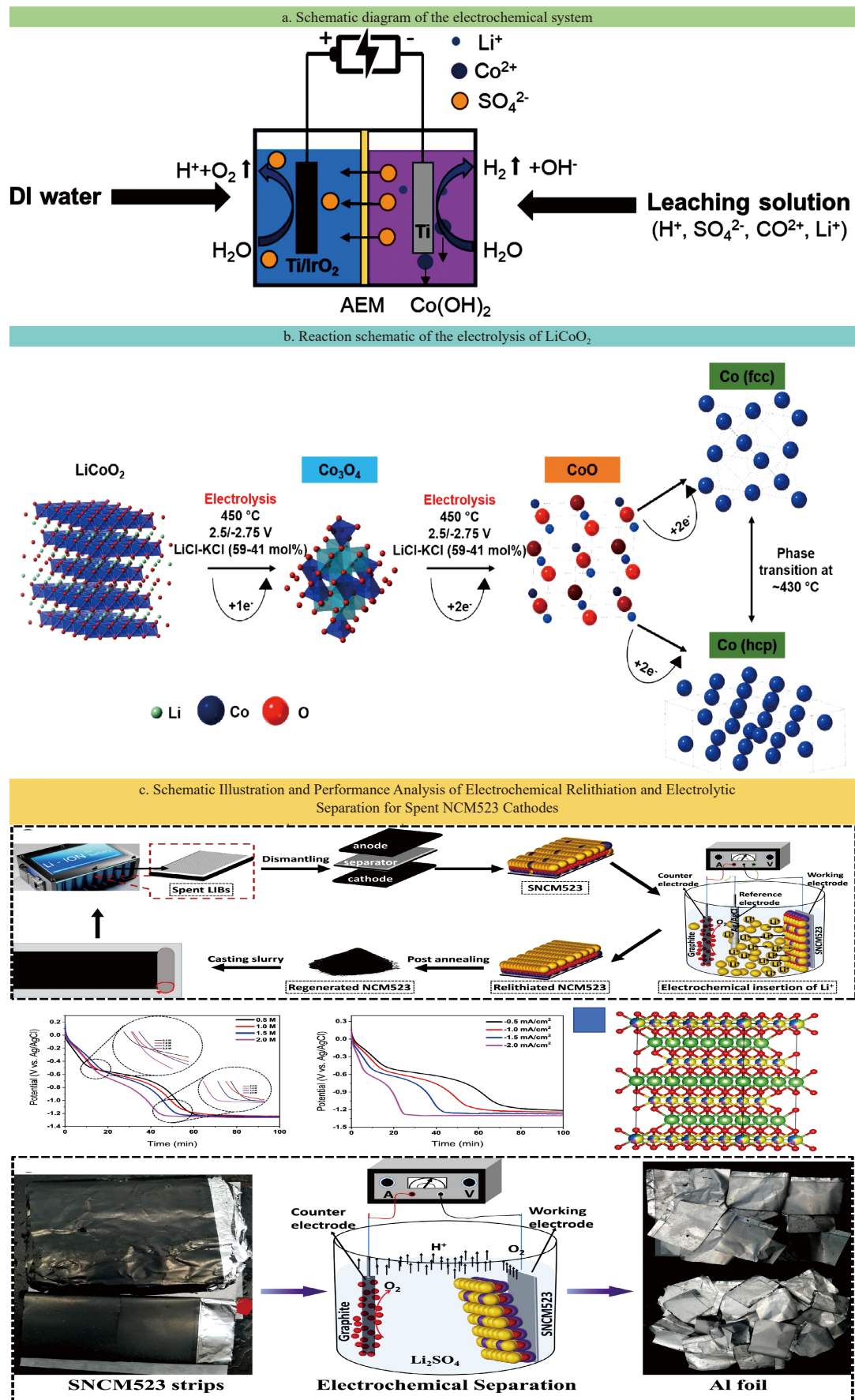
(e.g., Li^+)^[124].

Isaksson et al.^[125] proposed a three-stage electrodialysis process for recovering lithium and cobalt from LiCoO_2 (LCO) batteries. The core innovation involves utilizing EDTA to chelate cobalt, enabling the separation of Li^+ from $[\text{CoY}]^{2-}$ complexes through electrodialysis, followed by acid stripping and salt splitting steps to separately recover sulfuric acid, EDTA, CoSO_4 , and LiOH . Although this method demonstrates potential for reducing chemical consumption and by-product generation, further improvements are required regarding product concentration and current efficiency. Sun et al.^[126] developed an innovative “indirect electrochemical leaching” method for efficient recovery of lithium and cobalt from spent LCO batteries. As illustrated in Fig. 11a, this system regenerates sulfuric acid at the anode through electrochemical reactions for cyclic leaching, while simultaneously achieving Co^{2+} precipitation via pH control at the cathode, with Li^+ remaining in the solution. Experimental results demonstrate that under optimized conditions (0.25 M sulfuric acid, 80 °C, 30 min), both lithium and cobalt leaching efficiencies reached 100%, with sulfuric acid recovery exceeding 94% and stable performance maintained over 7 cycles. This approach significantly reduces acid-base consumption and wastewater discharge, thereby establishing a novel pathway for green recycling of LCO batteries. Mirza et al.^[127] constructed a customized micro high-temperature electrochemical cell integrated with in situ X-ray imaging technology, achieving the first direct real-time observation of LiCoO_2 electrochemical reduction to metallic cobalt in LiCl-KCl molten salt. As illustrated in Fig. 11b, the reduction pathway proceeds stepwise through intermediate phases of Co_3O_4 and CoO , with the final metallic Co product tending to adopt a face-centered cubic crystalline structure at temperatures above 450 °C. Through X-ray CT quantification of electrode structural evolution, cobalt metal deposition distribution, and gas evolution behavior, the study elucidates the correlation mechanism between electrode degradation and bubble dynamics in molten salt electrochemical recycling processes, thereby providing critical microstructural insights for optimizing molten salt battery recycling strategies. Feng et al.^[128] proposed an electrochemical approach for the direct recovery of cathode materials from spent LFP batteries. By employing a zero-gap membrane electrode assembly (MEA) reactor, lithium was efficiently extracted and directly converted into high-purity lithium hydroxide (LiOH) with low energy consumption (as low as 103 kJ kg^{-1}) and high product purity ($\approx 99\%$). Experimental results demonstrated stable operation of the 20 cm^2 reactor for over 1000 h, achieving a lithium recovery rate of 89.8% and a Faradaic efficiency of up to 96.4%. Moreover, this method exhibited applicability to various cathode materials, such as LiMn_2O_4 and NMC532, highlighting its scalability potential and industrial adaptability. This work offers a promising green and efficient pathway for recycling SLIBs. Yan et al.^[129] developed an oxidation- CO_2 leaching process for the efficient extraction of lithium and control of impurities from spent LiFePO_4 cathode materials. In this approach, LiFePO_4 was first oxidized to FePO_4 , followed by mild leaching using a

$\text{CO}_2\text{-H}_2\text{O}$ carbonic acid system, which avoids the use of strong acids and enables selective lithium dissolution while suppressing co-dissolution of impurities. Experimental results demonstrated a lithium leaching efficiency exceeding 95%, with significantly reduced concentrations of impurities such as iron and aluminum in the obtained solution. This provides a high-quality feedstock for subsequent production of high-purity lithium products. The process represents a green and low-energy strategy for the recycling of SLIBs. Mehboob et al.^[130] proposed a closed-loop strategy termed “electrochemical lithium compensation-stripping + molten salt repair” for the regeneration of spent NMC523 cathodes. As illustrated in Fig. 11c, the process first involves rapid (on the order of seconds) detachment of the aluminum foil in 2 M Li_2SO_4 via electrochemical stripping, followed by lithium replenishment, elimination of oxygen vacancies, and mitigation of cation mixing through treatment in a LiOH-NaCl molten salt at 850 °C. The regenerated material exhibits a crystal lattice ordering comparable to that of commercial counterparts, with an enhanced Li^+ diffusion coefficient of $7.46 \times 10^{-8} \text{ cm}^2 \text{ s}^{-1}$. Electrochemical testing shows a capacity retention of 78.2% after 1000 cycles at 1C. Furthermore, this approach reduces energy consumption and carbon emissions by 60%–75% compared to conventional pyrometallurgical or hydrometallurgical routes, achieving high-value direct regeneration of spent cathodes.

Current electrochemical recycling technologies for SLIBs are confronted with multiple critical bottlenecks: Firstly, inadequate recovery efficiency and insufficient product purity remain significant challenges, with particularly suboptimal metal recovery performance observed for novel complex electrode materials such as high-nickel, high-voltage, and solid-state batteries. Secondly, technologies such as electrodialysis suffer from low product concentration and unsatisfactory current efficiency, thereby resulting in poor economic viability. Thirdly, severe membrane fouling adversely affects ion migration and enrichment efficiency, thereby elevating operational costs. Fourthly, pronounced electrode degradation occurs during molten salt electrolysis, where anodic gas evolution significantly increases reaction resistance. Additionally, high recycling costs are highly susceptible to raw material price fluctuations, consequently dampening corporate adoption incentives.

Future research should prioritize the following directions: optimizing electrochemical processes to enhance electrodialysis concentration and current efficiency, mitigating membrane fouling, and improving molten salt electrolysis to delay electrode degradation; developing efficient recycling technologies compatible with next-generation battery systems; integrating multidisciplinary approaches encompassing physical and chemical routes to establish synergistic processes that improve recovery efficiency and purity while reducing costs; focusing on breakthroughs in efficient separation and purification of valuable metals while promoting closed-loop recycling of auxiliary substances such as sulfuric acid; and incorporating artificial intelligence and automation technologies to streamline sorting and logistics processes and enhance the value of recovered materials, thereby establishing economi-



cally viable pathways for large-scale implementation.

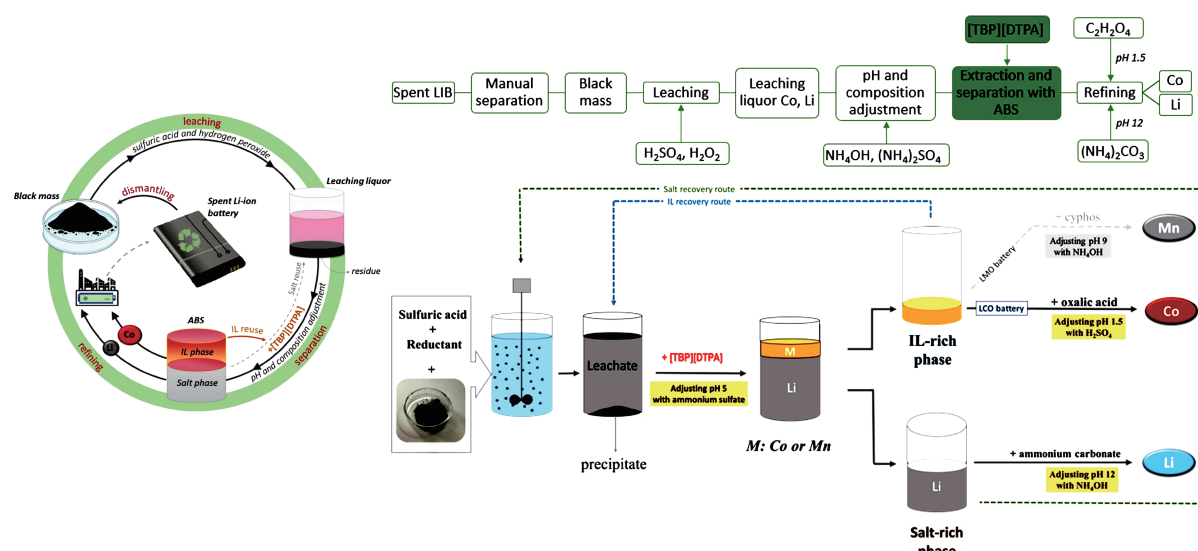
4.3. Ionic liquid-based recycling technologies

The pivotal foundation of ionic liquid-based recycling technologies resides in harnessing the distinctive physicochemical properties of ionic liquids, including their negligible vapor pressure, exceptional thermal stability, and tunable solvation capabilities^[131,132]. The recycling process primarily involves three key steps: First, the battery electrolyte composition is analyzed to identify target components. Subsequently, appropriately selected or customized ionic liquids are employed to dissolve the electrolyte, enabling its separation from the electrode materials. Finally, utilizing ionic liquids as leaching agents, valuable metals such as cobalt, nickel, and lithium within the electrode materials are dissolved, followed by their selective extraction and purification either leveraging the inherent properties of the ionic liquids or in combination with extractants, ultimately yielding high-purity metal products. This technology demonstrates significant multifaceted advantages compared to conventional methods. From an environmental perspective, ionic liquids exhibit negligible volatility, thereby preventing the release of hazardous gases, and the process operates without requiring large quantities of highly corrosive acids or alkaline solutions, significantly reducing the generation of dangerous liquid waste^[133]. Furthermore, their reactions typically proceed under relatively mild temperature conditions, contributing to lower energy consumption. In terms of efficiency, the powerful solvation capability of ionic liquids towards metals enhances metal recovery rates, while their favorable dissolution properties accelerate reaction kinetics, thereby shortening the overall recycling duration. Regarding adaptability, by tailoring the chemical structure and composition of ionic liquids, this technology can be effectively applied to batteries incorporating various cathode materials (e.g., NMC, LFP), demonstrating considerable potential for scaling from laboratory research to industrial implementation^[134].

Mušović et al.^[135] reported a mild aqueous biphasic system for the separation and recovery of valuable metals from spent LCO cathodes. The critical finding worth highlighting is that the ionic liquid maintained extraction efficiency above 87% after four cycles, but the study did not report whether impurity accumulation occurs over more extended cycling—a key unknown for industrial feasibility. After initial leaching with sulfuric acid and hydrogen peroxide, the [TBP][DTPA] ionic liquid was employed for one-step selective extraction, achieving a Co(II) recovery rate of over 95% into the ionic liquid phase while Li(I) remained in the brine phase. Subsequent precipitation steps using oxalic acid for cobalt and ammonium carbonate for lithium yielded the respective metal compounds. The ionic liquid maintained an extraction efficiency above 87% even after four regeneration cycles. As illustrated in Fig. 12a, the process features low energy and water consumption and can be seamlessly integrated with conventional sulfate-based recycling routes, offering a green and efficient pathway for Co/Li separation from SLIBs. Guo et al.^[136] constructed a synergistic extraction system utilizing the ionic liquid [APMIM][Tf₂N] and Cyanex 923, enabling selective extraction and recovery of cobalt, nickel, and man-

ganese from spent ternary LIBs, while retaining lithium in the raffinate. The study demonstrated that this system achieves highly efficient separation via a combined “cation exchange and coordination complexation” mechanism, with the extractant exhibiting excellent regenerability and stability. Jing et al.^[137] designed and synthesized a novel task-specific ionic liquid, [C₁₂H₂₅NH₃][Cyanex 272], for the efficient separation and recovery of V(V) from concentrated Cr(VI) solutions. Under optimized conditions (initial pH: 1.5, extractant concentration: 0.26 mol L⁻¹, extraction time: 25 min), a V(V) extraction efficiency of 99.5% was achieved, with a separation factor $\beta_{V/Cr}$ of $\sim 1.08 \times 10^3$. The loaded organic phase was subsequently washed with water and stripped with ammonia solution (0.52 mol L⁻¹ NH₃·H₂O, 40 min), attaining a stripping efficiency of 99.2%. Plate-like NH₄VO₃ crystals matching the reference XRD pattern were directly obtained after crystallization. The ionic liquid retained a recovery rate > 97% even after four consecutive cycles, demonstrating its excellent reusability. This process provides a green, highly selective, and low-volatility pathway for the clean separation of vanadium from chromium. Shamanth et al.^[138] have combined density functional theory (DFT) and molecular dynamics (MD) simulations to elucidate the lithium extraction mechanisms of four ionic liquids at the molecular level. By evaluating binding energies and electronic structures, [N₄₄₄₄][EHPMEH] was identified as exhibiting optimal Li⁺ binding affinity and selectivity, while MD simulations validated its high extraction performance through analysis of interfacial migration and diffusion coefficients. Chen et al.^[139] developed a green and efficient recycling strategy for all-solid-state sodium-ion batteries. Through systematic comparison of three green solvents, low-melting mixture solvents (LoMMSs) were identified as achieving the highest sodium leaching efficiency (92.8%) under mild conditions with excellent selectivity. The study further implemented anti-solvent screening to accomplish highly efficient sodium precipitation (92.0%), confirming the scalability of this methodology and thereby establishing a novel strategy for sustainable recycling of ASIBs. Do et al.^[140] utilized Nipa palm shell as a raw material to prepare microcrystalline cellulose (MCC) with a purity of 96.2% through an “alkali-acid-bleaching” process. The MCC was then phosphorylated in a one-step reaction using phosphoric acid, urea, and ammonium salts to obtain a Li⁺-selective biosorbent with high specific surface area and abundant PO₄³⁻ groups. Under optimized conditions (pH: 6, adsorbent dosage: 0.12 g mL⁻¹, contact time: 480 min) for a 10 mg L⁻¹ Li⁺ solution, the adsorption capacity reached 5.4 mg g⁻¹, with the process well described by the Freundlich model and pseudo-second-order (PSO) kinetics. In parallel, the task-specific ionic liquid [C₄H₉NH₃][Cyanex 272] was synthesized. At pH 3.5, within 15 min, and an organic-to-aqueous (O/A) ratio of 1:1 under three-stage cross-flow extraction, the separation efficiency between Ni²⁺ and Co²⁺ was enhanced from 33% to over 90%, achieving a Co²⁺ extraction rate of 93.6% while retaining only 1% of Ni²⁺ in the organic phase. As illustrated in Fig. 12b, this integrated “biosorption-ionic-liquid extraction” strategy enables the efficient one-step separation and recovery of Li⁺, Ni²⁺, and Co²⁺, offering a low-cost, low-emission, and green closed-loop solution for SLIB recycling.

a. Sustainable recovery of cobalt and lithium from lithium-ion battery cathode material by combining sulfate leachates and aqueous biphasic systems based on tetrabutylphosphonium-ionic liquids



b. Green efficient dual-strategy for critical metal recovery from spent LIBs: Nipa palm shell-derived cellulose and $[C_4H_9NH_3][\text{Cyanex 272}]$ ionic liquid extraction

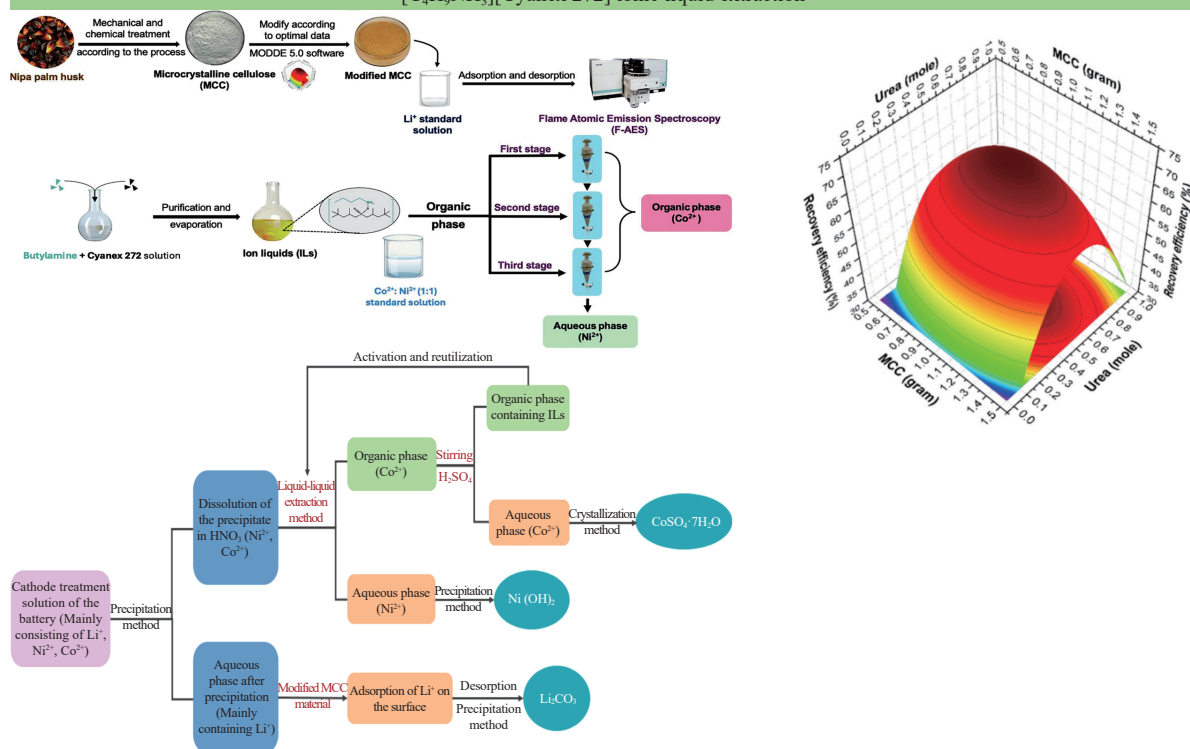


Fig. 12. Green recovery of critical metals (cobalt/lithium/nickel) from SLIB cathodes: Flowchart of ionic liquid+aqueous biphasic system/cellulose-based dual strategy. (a) Reproduced with permission from Ref. [135], (b) Reproduced with permission from Ref. [140].

The application of ionic liquid technology in the field of SLIB recycling still confronts multiple critical bottlenecks: First, cost constitutes the primary barrier, as the complex synthesis procedures and expensive raw materials for ionic liquids directly result in poor economic viability of the recycling process, hindering large-scale implementation. Second, the separation efficiency for critical metals remains unsatisfactory; although ionic liquids can dissolve various valuable met-

als, they exhibit low selectivity and suboptimal recovery rates for lithium ions. Specifically, lithium's high solubility in the aqueous phase makes efficient and short-range targeted separation challenging. Furthermore, mechanistic understanding remains inadequately elucidated; the complex interaction mechanisms between ionic liquids and different metal ions are still unclear, impeding precise process design and optimization. Finally, scale-up issues are prominent—transitioning

from laboratory to industrial scale presents challenges such as poor equipment compatibility and unstable process performance, which obstruct practical implementation of the technology.

To overcome the aforementioned challenges, future development of this technology should prioritize the following strategic directions: Firstly, advancing the rational design of functional ionic liquids through molecular engineering to develop novel systems that simultaneously exhibit high selectivity, superior stability, and reduced costs, thereby accommodating diverse battery materials. Secondly, deepening fundamental mechanistic investigations by leveraging advanced techniques such as density functional theory calculations and molecular dynamics simulations to systematically elucidate the microscopic mechanisms governing dissolution and separation processes, thus establishing a theoretical foundation for process development. Thirdly, promoting synergistic integration by combining ionic liquid-based methods with complementary technologies including pretreatment, charged disassembly, and direct regeneration to form holistic, efficiency-enhanced recycling solutions. Fourthly, strengthening engineering scale-up and establishing comprehensive closed-loop frameworks by addressing critical challenges in equipment design, energy consumption, and operational stability during large-scale implementation, while actively promoting the development of integrated “recycling-regeneration-reutilization” systems to ultimately achieve efficient resource circulation and minimized environmental impact.

4.4. Comparative analysis of emerging recycling technologies

Current emerging LIB recycling technologies are primarily represented by three major categories: direct regeneration and repair, electrochemical recycling, and ionic liquid-based recycling, each with distinct technical emphases, as summarized in Table 3. Direct regeneration and repair technologies are designed to restore cathode material performance to near-pristine levels through crystal structure reconstruction and lithium replenishment, enabling direct material reutilization. However, their high-temperature or high-pressure operations result in substantial energy consumption and stringent equipment requirements^[141]. Electrochemical recycling technologies recover high-purity metals from leachates via electrolysis, demonstrating exceptional recovery efficiency for cobalt and nickel with minimal environmental pollution, although they are constrained by high electricity demand and specific leachate composition requirements^[142]. Ionic liquid-based recycling, as an emerging methodology, utilizes ionic liquids to achieve preferential lithium extraction and precise cobalt separation, featuring rapid reaction kinetics, environmental compatibility, and high product purity. Nevertheless, it currently represents the most expensive pathway due to prohibitively high solvent costs and challenges in solvent regeneration^[143]. In summary, these three technological pathways demonstrate distinct trade-offs among recovered products, core advantages, cost structures, and environmental impacts. Their future development trajectories will hinge on breakthroughs in overcoming respective technical bottlenecks to optimize the balance

between economic viability and recycling effectiveness, thereby enhancing overall sustainability.

To advance scalable and commercially viable applications, these three technologies must focus on overcoming their core bottlenecks: Direct regeneration and repair should develop low-temperature/pressure green regeneration processes to reduce energy consumption, while advancing intelligent lithium compensation technologies to ensure performance consistency. Electrochemical recycling needs to enhance energy efficiency through optimized electrode and electrolyzer design. Ionic liquid-based recycling requires the development of novel solvents with lower costs and higher stability, alongside establishing effective solvent circulation systems to control operational expenses. Furthermore, promoting the coupling and integration of these technologies to form hybrid processes represents a crucial trend toward achieving efficient, clean, and economically sustainable recycling.

5. Industry landscape and policy implications

The recycling of SLIBs has rapidly evolved from laboratory-scale research into a policy-driven industry of significant scale. The entire recycling chain constitutes a closed-loop process, encompassing the collection of spent batteries, safe disassembly, crushing and sorting, and the extraction of regenerated materials via hydrometallurgical/pyrometallurgical processes or direct regeneration technologies, with subsequent reintroduction into production, as illustrated in Fig. 13a. A fundamental distinction exists between industrial and laboratory-scale processes within this chain: the former prioritizes large-scale operation, economic viability, and process stability while managing complex and highly heterogeneous feedstocks; the latter focuses on proof-of-principle validation and exploration of performance limits, generating fundamental knowledge and high-purity samples, as depicted in Fig. 13b. A cost/energy consumption heatmap reveals critical bottlenecks (Fig. 13c): hydrometallurgy emerges as an economic hotspot due to high reagent and pollution control costs, while pyrometallurgy constitutes an energy consumption hotspot owing to its high-temperature smelting processes. Together, these factors constrain commercial viability. In contrast, direct regeneration shows significant potential in both energy and cost savings, yet its practical application is currently limited by stringent feedstock pretreatment requirements. As illustrated in Fig. 13d, the environmental impact infographic highlights challenges across various stages: hydrometallurgical processes generate chemical wastewater, pyrometallurgical routes emit greenhouse gases and flue gases, while direct recycling exhibits the potential for the lowest environmental footprint. Consequently, global policies are systematically guiding technology selection, capacity allocation, and business models through mandatory regulations, economic incentives, and industrial planning. This coordinated approach drives the industry toward an optimal balance among resource recovery efficiency, economic viability, and environmental friendliness, thereby establishing a truly sustainable lifecycle management system for batteries (Fig. 13e).

The European Union's Battery Regulation stands as one of the most forward-looking and systematic policy frame-

Table 3. Comparative analysis of core indicators for three emerging resource recycling technologies for SLIBs^[141-148].

Aspect	Direct regeneration & repair	Electrochemical recycling	Ionic liquid-based recycling
Core principle	Restores electrochemical performance by repairing cathode lattice structures and replenishing lithium loss via physical or chemical methods.	Dissolves spent cathode material in acid, followed by electrodeposition of valuable metals at the cathode.	Utilizes ionic liquids as solvents to selectively dissolve and separate valuable metals from spent batteries by tuning composition and reaction conditions.
Key advantage	Directly yields cathode material with performance comparable or superior to conventional synthesis without destroying the original crystal structure.	Enables efficient enrichment of valuable metals with minimal pollution and reduced risk of introducing new impurities.	Allows preferential lithium extraction and precise cobalt separation without extra reductants/precipitants, features fast kinetics and low environmental risk.
Main limitation	High energy consumption for high-temperature solid-state methods, stringent purity requirements for feedstocks, and hydrothermal methods require high-pressure equipment with strict safety specifications.	High electricity consumption and specific requirements for leachate composition.	Relatively high cost of ionic liquids; regeneration and reuse of certain types remain challenging.
Metal recovery rate	Effectively retains metallic elements in cathode materials, achieving material-level regeneration.	High recovery efficiency for common metals such as Co and Ni.	Particularly high lithium recovery efficiency; some routes exceed 90%.
Product purity/performance	Regenerated cathode performance can be restored to near-pristine levels.	High-purity metals can be obtained through precise control of electrolysis and purification steps.	Customizable ionic liquids enable precise separation; black mass purity can exceed 95%.
Typical energy consumption (kWh/kg black mass)	High: 200–500 (mainly from high-temperature solid-state sintering)	Moderate to high: 150–400 (electrolysis is the primary energy-consuming step)	Low-moderate: 50–150 (operated at ambient or low temperature, with energy mainly used for mixing and separation)
CO ₂ emissions (kg CO ₂ /kg black mass)	Moderate to high: 100–300 (high for solid-state sintering, lower for hydrothermal routes)	Moderate: 50–150 (depends on electricity source, no direct emissions from electrolysis itself)	Low: 20–80 (low-temperature, non-volatile processes, emissions mainly from reagent production and electricity)
Processing cost (USD/t spent batteries)	Moderate: 500–1200 (high energy cost for solid-state, high equipment/operational cost for hydrothermal)	Moderate: 600–1500 (primarily electricity and electrode costs)	High: 1000–3000 (high cost of ionic-liquid raw materials is the main factor)
Processing time	Short to moderate: 1–3 d (solid-state sintering). Hours to 1 d (hydrothermal reaction, though pre-/post-treatment extends overall cycle)	Moderate: 1–5 d (including leaching, electrolysis, and post-processing)	Moderate: hours to 2 d (fast dissolution/separation, but ionic-liquid regeneration may increase total time)
Equipment requirement	High (precise sintering furnaces or high-pressure reactors, stringent temperature/pressure control)	Moderate (electrolytic cells, power supply, and solution-management systems)	High (corrosion-resistant sealed reactors and efficient ionic-liquid recovery/purification systems)
Environmental impact	High carbon emissions for solid-state routes, relatively low for hydrothermal methods.	Electrolysis itself is low-pollution, but the preceding acid-leaching step may generate wastewater.	Environmentally benign—ionic liquids exhibit negligible emissions of volatile organic compounds and low toxicity.
Scalability assessment	Moderate—Strength: well-defined process, partial compatibility with existing cathode production lines; Challenge: strict feedstock consistency/purity requirements, difficulty in securing large-scale, stable waste supply, high energy-efficiency challenges for thermal routes.	Moderate—Strength: relatively compact flow, high product purity; Challenge: sensitivity to high electricity costs, electrolysis requires stable feed composition, power-optimization is key for scale-up.	Currently low—Strength: high separation selectivity, strong potential for green chemistry; Challenge: high ionic-liquid cost, immature closed-loop regeneration/recovery, significant economic barriers to large-scale adoption.
Applicable battery types	Best suited for structurally stable, easily relithiated single-component cathodes such as LFP and LCO.	Suitable for soluble, high-value-metal cathodes like NMC/NCA and LCO.	Potentially applicable to various LIB cathode materials, with unique advantages in selective Li extraction and separation of similar metals (e.g., Co/Ni).

works globally^[149]. It not only sets phased, mandatory targets for the recovery rates of critical metals such as cobalt, nickel, and lithium, but also introduces innovative requirements such as the “battery passport” and full life-cycle carbon footprint accounting^[150]. These provisions extend environmental responsibility to battery producers, mandating supply chain transparency and eco-design, thereby promoting the development of easily recyclable battery structures and the establishment of closed-loop material cycles from the out-

set. While digital tools such as battery passports and blockchain have been proposed to enhance traceability, their integration with physical recycling processes remains nascent, and claims of “intelligent” systems often outpace demonstrated metallurgical performance. Future progress will depend more on advances in separation chemistry, process engineering, and system integration than on AI alone^[151]. Its influence extends beyond Europe, serving as a key reference point for the global battery industry's green transition.

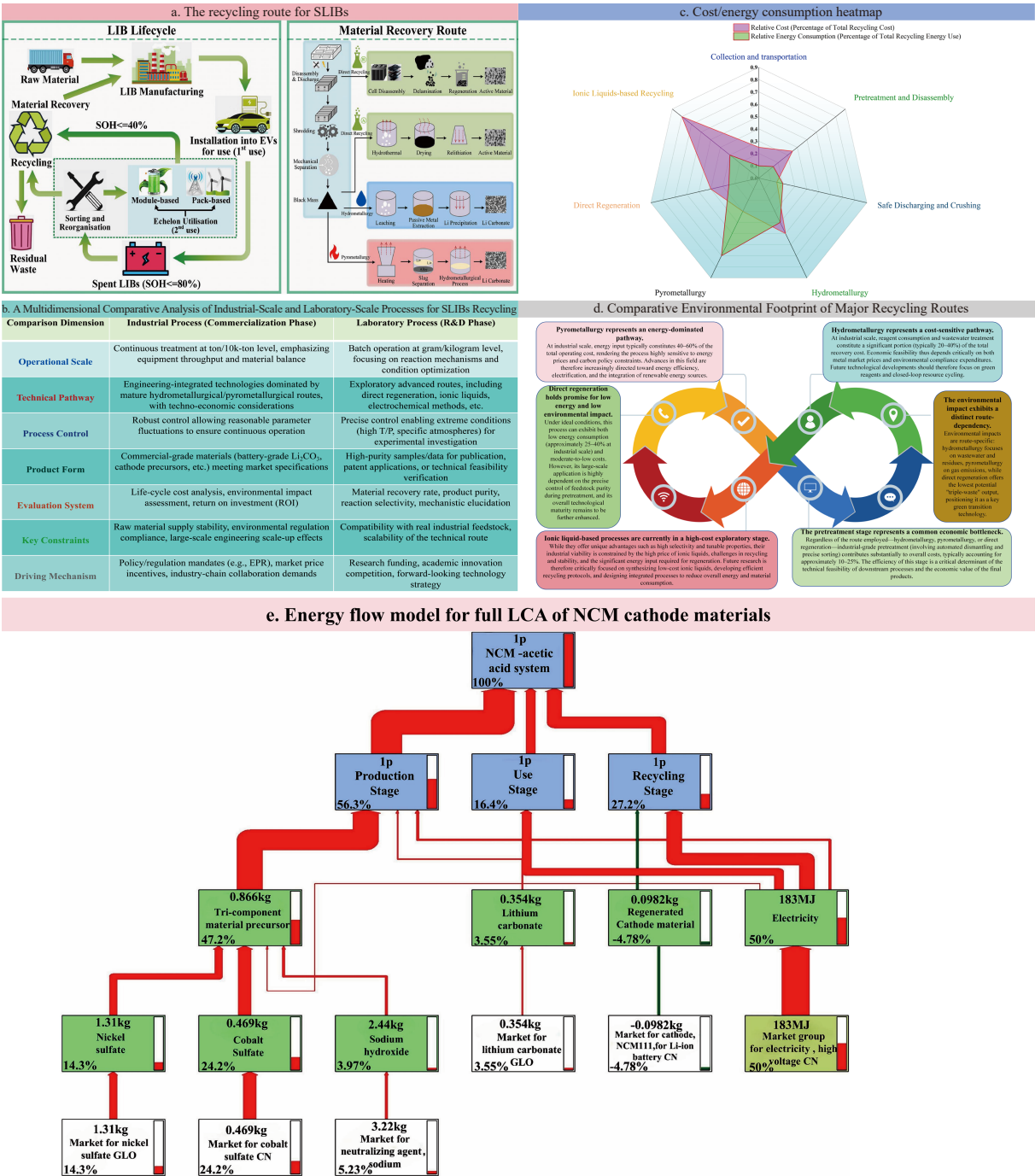


Fig. 13. Multidimensional constraints and system optimization in recycling spent lithium-ion batteries: Integrating technical, economic, and environmental perspectives. (a) Reproduced with permission from Ref. [52], (e) Reproduced with permission from Ref. [158].

The U.S. Inflation Reduction Act (IRA) exerts a decisive influence from the perspectives of market demand and supply-chain security^[152]. Through economic instruments such as tax credits, the Act explicitly mandates that a defined proportion of critical minerals contained in electric vehicle batteries must be extracted, processed, or recycled within North America or in countries with which the United States has a free trade agreement. This policy has significantly stimulated investment in battery recycling and material remanufacturing capacity within the United States and among its allied nations, accelerating the restructuring of regional supply chains. Consequently, it drives recycling technology development to simulta-

neously meet the demands for high efficiency and low cost, aiming to produce high-purity recycled materials that can be directly reintegrated into mainstream battery manufacturing supply chains. This, in turn, reinforces the demand-oriented push for technologies such as short-process and high-value direct regeneration^[153].

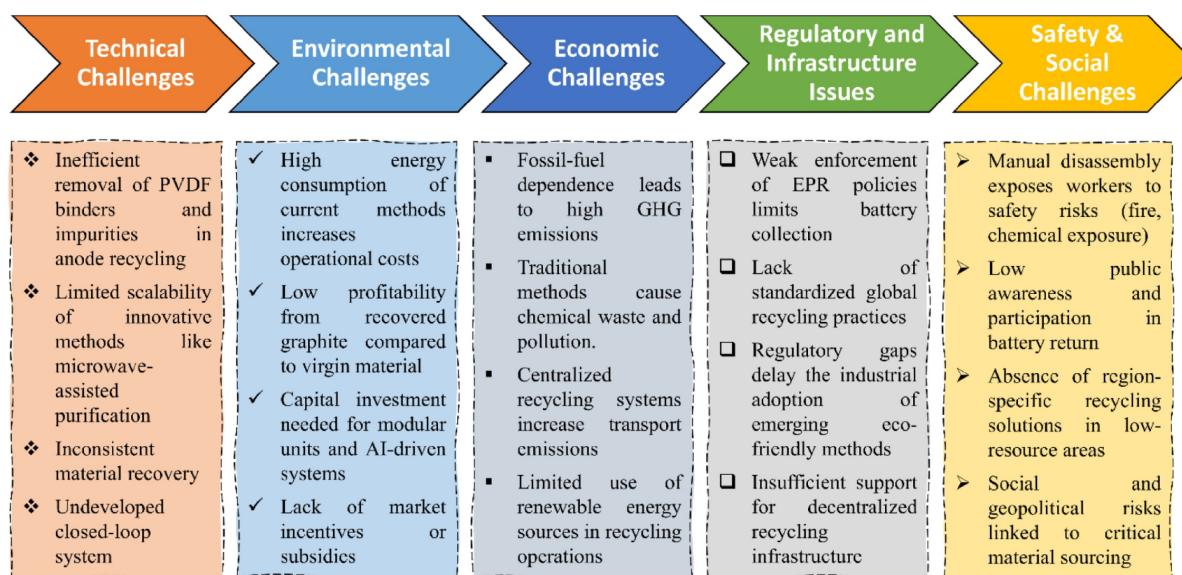
As one of the world’s largest producers and consumers of LIBs, China has established a policy and regulatory framework centered on the Extended Producer Responsibility system and a nationwide traceability management platform^[154]. By implementing a “White List” mechanism to standardize the qualifications of recycling enterprises and setting clear

recycling targets, China is promoting a dual-track pathway that prioritizes cascade utilization and ensures regenerative recycling^[155]. In response to the substantial market stock of LFP batteries, China has rapidly expanded its recycling capacity and is actively exploring market-driven incentives such as recycling quotas and carbon emission trading mechanisms. These measures aim to foster the growth of a compliant recycling ecosystem and facilitate the adoption of high-value recovery technologies^[156].

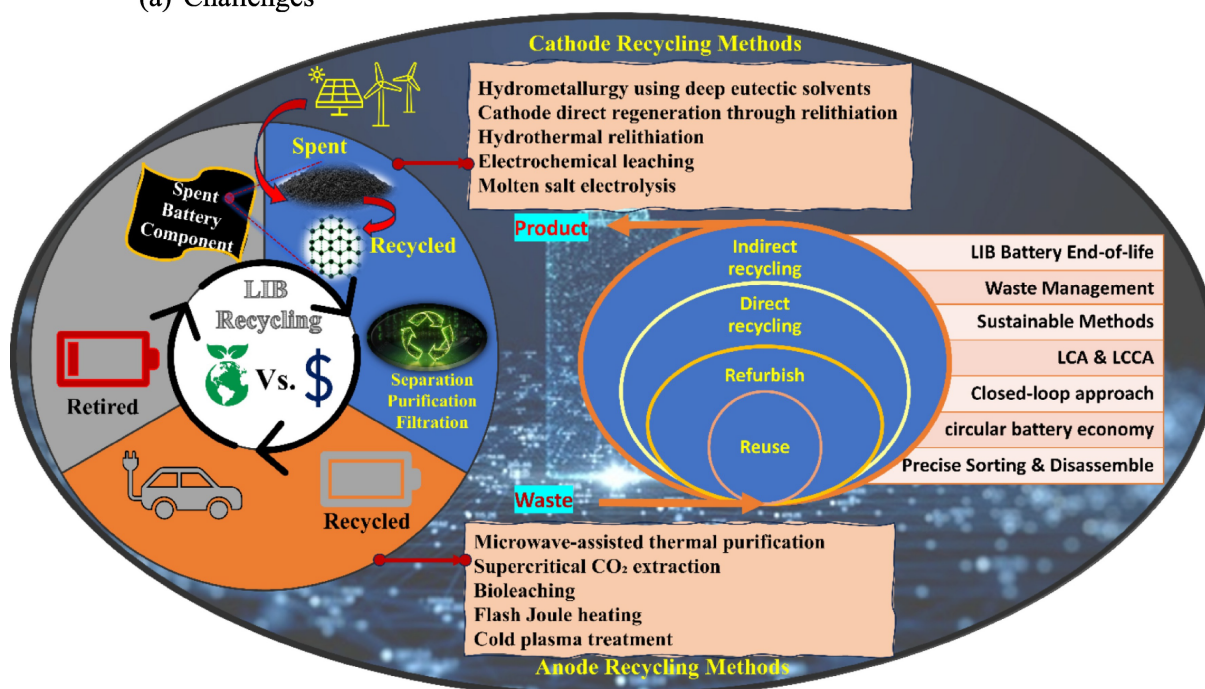
From the perspective of global recycling capacity trends, in response to the projected global volume of end-of-life power batteries exceeding 5 million metric tons by 2030, China, Europe, and North America are emerging as the three key regions for the development of core recycling capacity^[157]. However, the industry as a whole still faces struc-

tural challenges: the geographical distribution of high-quality recycling capacity remains uneven; technological pathways compete between the dominance of hydrometallurgy, the economies of scale of pyrometallurgy, and the industrial breakthrough of emerging green technologies; the lack of unified quality standards and certification systems for recycled materials restricts their large-scale application in downstream battery manufacturing; and insufficient integration of value chains and data sharing among battery production, vehicle manufacturing, and recycling stages affects the efficiency of closed-loop systems.

Looking ahead, the interaction between policy and industry is expected to exhibit three major trends, as illustrated in Fig. 14. First, increasingly stringent regulations will continue to drive recycling technologies toward greener, lower-carbon,



(a) Challenges



(b) Proposed recommendations

Fig. 14. Recommendation for future spent lithium-ion battery recycling. Reproduced with permission from Ref. [159].

and more intelligent solutions. Second, methodologies such as battery passports and carbon footprint accounting are likely to become key areas for international cooperation and mutual recognition of technical standards. Third, innovative business models such as “battery-as-a-service” and material banking will converge with advanced recycling technologies, collectively enhancing the economic and environmental sustainability of batteries throughout their lifecycle. Ultimately, these developments will propel the global battery industry toward a true circular economy system.

6. Conclusions

(1) Current pretreatment technologies are confronting scaling bottlenecks. Unlike existing reviews that focus on single pretreatment steps, this work reveals a systemic gap between laboratory efficiency and continuous-line stability. Future development must evolve toward continuous, well-instrumented, and process-integrated systems to enhance energy efficiency and minimize secondary pollution.

(2) Hydrometallurgical, pyrometallurgical, and bioleaching technologies each possess distinct advantages, yet they commonly confront challenges such as high energy consumption, suboptimal metal recovery rates, and environmental concerns. A key insight distinguishing this review is that these three mainstream routes share a common “feedstock sensitivity” bottleneck rather than independent limitations. Future progress requires multidisciplinary integration and systematic optimization to achieve efficient, low-carbon, closed-loop recycling systems.

(3) Direct regeneration and repair technologies enable direct recovery of battery performance. However, their large-scale application is constrained by limited remediation efficiency, stringent process control requirements, and high equipment costs. Future efforts should focus on low-temperature hydrothermal routes and intelligent lithium compensation as already demonstrated in the literature reviewed herein.

(4) Electrochemical recycling technologies achieve metal separation through electrodeposition or electrodialysis, offering high recovery efficiency and product purity under optimized laboratory conditions. However, industrial-scale application is constrained by membrane fouling, electrode degradation, high energy consumption, and leachate composition sensitivity. Future efforts should focus on integrated electrodialysis-electrodeposition systems and durable membrane/electrode materials to improve current efficiency and long-term stability.

(5) Ionic liquid technologies possess inherent advantages such as low volatility, high thermal stability, and structural tunability. However, their industrial implementation is hindered by prohibitively high synthesis costs, unsatisfactory recovery efficiency for target metals, and complex reaction mechanisms. Future development should concentrate on task-specific ionic liquid design and closed-loop solvent circulation to improve recyclability.

Acknowledgements

The authors received funding from the National Natural Science Foundation of China (51604118), Open Foundation of

State Key Laboratory of Mineral Processing Science and Technology (BGRIMM-KJSKL-2024-11), Science Research Project of Hebei Education Department (ZD2020132), Central Guide Local Science and Technology Development Project of Hebei (236Z4104G), The Hebei Natural Science Foundation (E2025209056), and Key Technological Innovation Projects of HBIS Group (HG2026115). Thanks for the aforementioned funding support.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Author contribution statement

Jianqiu Qin: Writing – original draft, Writing – review & editing, Investigation, Methodology, Visualization. **Shaoying Li:** Conceptualization, Funding acquisition, Supervision, Resources. **Beining Liu:** Data curation, Formal analysis, Software. **Xiaodong Yu:** Validation. **Liucheng Zhao:** Funding acquisition, Project administration, Supervision.

Use of AI statement

None.

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Jianqiu Qin is a Master's candidate in the College of Mining Engineering at North China University of Science and Technology. He is co-advised by Associate Professor Shaoying Li and Associate Professor Liucheng Zhao. His current research focuses on efficient separation processes and theory for complex refractory minerals, as well as the comprehensive utilization of secondary resources. He has published more than 10 papers in domestic and international journals such as *Materials Reports* (Chinese), *The Chinese Journal of Nonferrous Metals* (Chinese), *Green and Smart Mining Engineering*, and *Minerals Engineering*.



Shaoying Li received his MS degree from China University of Mining and Technology, Beijing, and his PhD from the University of Science and Technology Beijing. He is currently an Associate Professor at the College of Mining Engineering, North China University of Science and Technology. His research interests lie in the efficient separation of complex refractory minerals and the comprehensive utilization of secondary resources.



Liucheng Zhao earned his PhD from the University of Science and Technology Beijing, where he completed a combined master's and doctoral program. He is currently an Associate Professor at the College of Mining Engineering, North China University of Science and Technology. His research focuses on the efficient separation and comprehensive utilization of complex refractory minerals, the resource utilization of solid waste, and the value-added processing of non-metallic minerals.